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Letter**

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March 1996

**Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830**

**Pacific Northwest National Laboratory
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INTRODUCTION

Preliminary predictions were made of the time to reach hypothesized operational limits of the HWVP melter due to build up of a noble metals sludge layer on the melter floor. These predictions will be updated in the preliminary and final Melter Performance Assessment (MPA) reports due in FY 1994. Predictions were made with the TEMPEST computer program, Version T2.9h, for use in the MPA activity in the Pacific Northwest Laboratory's (PNL) Hanford Waste Vitrification Plant (HWVP) Technology Development (PHTD) effort.

The TEMPEST computer program (Trent and Eyler 1993) is a PNL-MA-70/Part 2 - Good Practices Standard (QA Level III) research and development software tool. Software Control Procedures (SCP-70-312) requires the following statement to be made:

Results are based on the use of unverified software. No assurance is expressed or implied as to the accuracy, completeness, or usefulness of this information.

This statement applies to all results presented herein. As part of the FY 1993 MPA activity, the proper documentation has been compiled to prepare the TEMPEST code for validation and verification that would lead to QA qualification. It is suggested here that this effort be continued in the future so that the above QA clause can be removed from all TEMPEST results.

HYPOTHESIZED LIMITING SCENARIOS

Several scenarios are hypothesized as limiting the lifetime of the HWVP melter in this analysis. These include:

- I. Assuming the design condition upper-to-lower electrode power ratio is 55/45:
 - A. The noble metals layer blocks the path to the discharge section such that glass cannot be poured from the melter.
 - B.1 $T_{\text{glass}} < 1050^{\circ}\text{C}$ as measured by the lower thermocouples (TCs) in the thermowell, or
 - B.2 The current requirement to either of the electrode pairs reaches the limit of the silicon controlled rectifier (SCR), or
 - B.3 $T_{\text{max}} > 1350^{\circ}\text{C}$
- II. Assuming skewing of power to the two electrode sets such that the upper TCs stay between 1125°C and 1175°C :
 - A. The noble metals layer blocks the path to the discharge section such that glass

cannot be poured from the melter.

B.1 $T_{\text{glass}} < 1050^{\circ}\text{C}$ as measured by the lower TCs in the thermowell, or

B.2 The current requirement to either of the electrode pairs reaches the limit of the SCR,
or

B.3 $T_{\text{max}} > 1350^{\circ}\text{C}$

RELATED WORK

The basis for the methodology used in the present HWVP melter analysis is in Grünwald et al (1993), Cooper et al (1993), and Anderson et al (1992). These are the Engineering-Scale Melter (ESM), the Research-Scale Melter (RSM), and the Gradient Furnace Test (GFT) reports, respectively. The ESM report contains the principal details and analysis methodology used in the HWVP melter computer model analysis. Figure 1 presents one of the principal results - agreement of predicted Ru material balance and retention with measured ESM test data. Discussion of these results and details of the computational methodology developed to obtain them is included in the ESM report.

ASSUMPTIONS

Assumptions made in these preliminary HWVP melter computer model calculations include:

- The melter operates with continuous pouring of glass. This was assumed in these preliminary analyses to provide a base of comparison of sludge layer build up as a function of amount of glass poured.
- The melt temperature as indicated by two control thermocouples is maintained at 1150°C unless the electrical limitations of the facility (transformer and SCR power or current limitations) are reached.
- The target noble metals concentrations in the glass are 0.113 wt% for RuO_2 , 0.024 wt% for Rh, and 0.029 wt% for Pd. These were determined from noble metals concentrations in current reference HWVP feed¹. These values were adjusted in the computer model source terms to account for an assumed 20 hours of pouring and 1 hour of feeding without pouring for each canister cycle.

¹ Memo M. L. Elliott to L. L. Eyler, "Noble Metals Concentrations in Reference HWVP Feed", June 3, 1993 which references: 1) Reference letter from D. E. Larson to Distribution, "Reference NCAW Feed for HWVP", February 17, 1993, and 2) Reference letter from D. E. Larson to R. A. Smith, "HWVP Feed and Glass Composition Revision Technical Note", May 13, 1991.

- The electric property variations are dominated by the "hairball" character of the RuO_2 particles although the settling of all three noble metals was modeled. The "hairball" RuO_2 formations have a dominant effect on the electric properties because relative small quantities of RuO_2 in a "hairball", by volume, result in large increases in electric conductivity.
- Sludge layer buildup is slow relative to the system time constant for thermal response. This assumption allows preliminary analysis of electrical and thermal conditions from steady, point conditions.
- SCRs are limited to 300 KVA (3000 Amps on 100 V taps; 2000 Amps on 150V taps; and 1000 Amps on 300 V taps - Reference Fluor Daniel dwg. SK-8457-E-001).

RESULTS

Preliminary results used to estimate time-to-failure are included in Figures 2 to 4. In Figure 2, the RuO_2 material balance is presented as a function of glass discharged for an assumed continuous pour rate of 100 kg-glass/hr for an assumed reference glass density of 2170 kg-glass/ m^3 at 1150°C. A continuous RuO_2 source rate of 0.1076 kg- RuO_2 /hr was used in these calculations. This rate was determined based on a preliminary assumption that there would be 20 hr of pouring followed by 1 hr of feeding without pouring during canister changeout, or $(0.113 \text{ wt\%-target-RuO}_2) = [0.113 \text{ kg-RuO}_2/\text{hr}]/(100 \text{ kg-glass/hr})$; $(0.113 \text{ kg-RuO}_2/\text{hr}) \cdot (20 \text{ hr-pour} / (20 \text{ hr-pour} + 1 \text{ hr-feed})) = 0.1076 \text{ kg-RuO}_2/\text{hr}$. This adjustment is required in the computer model to account for the computer model treating the melt as a constant volume. In Figure 2, the time integrated retention $[(\int M_{\text{RuO}_2, \text{in}} - \int M_{\text{RuO}_2, \text{out}})/(\int M_{\text{RuO}_2, \text{in}})]$ is 16.2% at 60 days. The amount of material retained in the melter includes mass in the glass and mass on the floor in a sludge layer. Total RuO_2 mass in the glass has reached an equilibrium value of 3.48 kg- RuO_2 which corresponds to $(2.02 \text{ kg-RuO}_2/\text{m}^3)/(2170 \text{ kg-glass/m}^3) = 0.093 \text{ wt\%}$ in the glass being poured. This equilibrium value compares to a target value of 0.113 wt% based on noble metal concentration for current reference HWVP feed and a target value of 0.1076 wt% for the RuO_2 source conditions in the computer model. These target values are the expected values if no retention occurs. However, due to particle settling, mass accumulation in the sludge layer on the floor continues to grow at a rate of 0.37 kg/day after the equilibrium condition is reached in the glass. This growth rate is directly proportional to the equilibrium concentration in the glass and the settling rate of the RuO_2 particles.

The growth of the sludge layer thickness, $H(t)$, based on the mass accumulation rate on the floor is presented in Figure 3. The thickness of the sludge layer as a function of time is calculated as

$$H(t) = M_{\text{RuO}_2, \text{Floor}}(t) / (\rho_{\text{RuO}_2} * A_{\text{Floor}} * C_{v, \text{max}}) \quad (1)$$

where

$H(t)$	= Thickness of sludge layer from the bottom, center of the melter
$M_{\text{RuO}_2, \text{Floor}}(t)$	= Mass of RuO_2 on the floor, kg
ρ_{RuO_2}	= Density of RuO_2 = 6970 kg/m ³
A_{Floor}	= Area of the floor in the computer model = 1.669 m ²
$C_{v, \text{max}}$	= Maximum packing volume fraction = 0.06
t	= Time.

The time rate of change of sludge layer depth is calculated by differentiating the above equation with respect to time to determine that $dH/dt = 0.053$ cm/day, based on the accumulation rate of 0.37 kg/day. Given that the reference distance from the bottom of the HWVP melter to the lower edge of the lower electrode is 3.4 in. (8.6 cm), it would take $(8.6 \text{ cm}) / (0.053 \text{ cm/day}) = 162$ days at 100% operating efficiency for the sludge layer to build up to the lower edge of the lower electrodes. The distance to the center of the pour spout is $(3.4 + 8.75) = 10.9$ in. = 27.68 cm. The time for the sludge layer to reach this depth is $(27.68 \text{ cm}) / (0.053 \text{ cm/day}) = 522.3$ days = 1.43 years at 100% operating efficiency.

Figure 4 presents the electric and thermal results as a function of sludge layer depth measured from the bottom of the melter. Two sets of results are included: One set of results (open symbols) are for the situation where total power is maintained constant to keep the control thermocouple temperatures approximately equal to 1150°C. In this case, a current limit of 3000 Amps is imposed on the lower electrode pair and the power to the upper electrode pair is allowed to exceed the 300 KVA limit to maintain the 1150°C target temperature. The second set of results (solid symbols) in Figure 4 are for the situation where the 3000 Amp current limit is imposed on the lower electrode pair and the 300 KVA limit is imposed on the upper electrode pair.

For no sludge layer, the upper-to-lower power ratio is $234.2 \text{ kW} / 191.4 \text{ kW} = 1.22 = 55/45$. This ratio remains constant at a sludge layer depth of 5 cm. However, electric current required to maintain the power level at a point where the target temperature of 1150°C is maintained has increased significantly. The required increase in power is due to shorting effect of the sludge layer.

A significantly noticeable effect of the sludge layer is in the predicted temperatures. At 5 cm depth, the electrodes are not completely bridged, but electric current concentration has occurred such that local heating in the thin region just under the lower electrode pair results in significantly increased temperature. The predicted maximum temperature near the edge of the lower electrode, $T_{\text{max}, B}$, of 1325°C is near enough to the postulated failure limit II.B.3 to be of concern. By the time the sludge layer reaches the lower electrode (7.6 cm in the computer model, 8.6 cm in HWVP), the current in the lower electrode pair has reached the 3000 Amp limit and power in the upper electrode pair has reached the 300 KVA limit. From this point on, total power decreases and the melt temperature decreases also. The

failure limit II.B.1 ($T_{\text{glass}} < 1050^{\circ}\text{C}$) is reached at a sludge depth of 14 cm. At a growth rate of 0.053 cm/day, this limit is reached in $(14 \text{ cm}) / (0.053 \text{ cm/day}) = 264 \text{ days} = 0.72 \text{ years}$ at 100% operating efficiency.

SUMMARY OF ESTIMATED TIMES TO REACH FAILURE

The following summarizes the preliminary estimated time to reach the postulated failure modes for sludge layer depth growth rate of 0.053 cm/day resulting from noble metals feeding conditions corresponding to a continuous glass pour rate of 100 kg/hr and an RuO_2 source rate of 0.1076 kg- RuO_2 /hr. Time estimates are for 100% operating efficiency and are estimated in terms of days-of-pouring, not real time, for reasons discussed later.

I. Occurrence of deviation from 55/45 power ratio - Estimate: Prior to 162 days-of-pouring.

- A. Discharge blocked - (Not determined, but greatly in excess of other limits).
- B.1 $T_{\text{glass}} < 1050^{\circ}\text{C}$ - Estimate: 264 days-of-pouring.
- B.2 Lower pair SCR limit reached - Estimate: 162 days-of-pouring.
- B.3 $T_{\text{max}} > 1350^{\circ}\text{C}$ - Estimate: 1325°C reached in 94 days-of-pouring.

II. Assume power skew deviation to maintain $1125^{\circ}\text{C} < T_{\text{melt}} < 1175^{\circ}\text{C}$.

- A. Discharge blocked - (Not determined, but greatly in excess of other limits).
- B.1 $T_{\text{glass}} < 1050^{\circ}\text{C}$ - Estimate: Does not occur.
- B.2 SCR limits exceeded - Estimate: 162 days-of-pouring lower pair exceeds 3000 Amps.
Estimate: 162 days-of-pouring upper pair exceeds 300 KVA.
- B.3 $T_{\text{max}} > 1350^{\circ}\text{C}$ - Estimate: 1325°C reached in 94 days-of-pouring.

Variability in these preliminary time estimates to reach postulated failure limits are estimated to be within a range of a factor of 2 under estimated to a factor of 4 over estimated. The following discussion presents the basis for this range.

DISCUSSION

The above time estimates are for 100% operating efficiency and continuous pouring. Thus, the times are estimated as days-of-pouring, not real time. The time estimates could be approximately adjusted for operating efficiency by dividing the time estimate in days-of-pouring by the operating efficiency. HWVP design operation efficiency is 70%. Such a simple adjustment is not strictly correct, however, because it would not account for sludge layer depth increase that would occur during idle times. For example, at 70% efficiency,

there would be $(1.0 - 0.7) * 365 \text{ days} = 109.5 \text{ days}$ of idling per year. During idling, the maximum increase in sludge layer depth is limited to total settling of the amount of noble metals in equilibrium in the glass. Preliminary analysis shows this to be approximately 0.5 cm at a maximum packing concentration of 6 vol%. 30 days of continuous idling leads to settling of approximately 95% of the material. By comparison, 30 days of continuous pouring results in preliminary predicted sludge layer thickness of $30 \text{ days} * 0.053 \text{ cm/day} = 1.59 \text{ cm}$. Thus, adjusting the above days-of-pouring time estimates for operating efficiency requires that assumptions be made about the number and duration of idling periods. Adjustment for, and analysis of, these types of effects are expected to be part of the final Melter Performance Analysis report.

Consideration of a design-point condition of continuous slurry feeding in conjunction with 20 hours of pouring and 1 hour of feeding without pouring while changing canisters could be expected to affect these analyses by at most a ratio of the feeding-without-pouring time to total time $[1 \text{ hr} / (20 \text{ hr} + 1 \text{ hr})] = 4.7\%$. Note that as the feeding-without-pouring time increases, the effect would be increased until equilibrium is re-established by increased pouring rates after canister changing.

There are uncertainties in these preliminary estimates. The analysis procedure was developed and validated against the ESM retention data. Thus if it is assumed that the ESM retention data are accurate (Figure 1), the HWVP computer model retention results should be reasonably accurate (Figure 2). Uncertainties in the sludge layer build up rate can then be assumed to be only a function of parameters affecting sludge layer depth which are not directly involved in retention. Of these, the one with the largest uncertainty is the maximum packing fraction, $C_{v,max}$. A value of 0.06 ($= 6 \text{ vol}\%$) was used in the present analysis. This value was determined by visual examination and analysis of transmitted and reflected light micrographs of RSM and ESM glass samples. 6 vol% corresponds to 19 wt% of 6970 kg-RuO₂/m³ material in 2170 kg/m³ glass. This value is in reasonable consistency with Krause and Luckscheiter (1991) noble metals data which were at a maximum of 16.5 wt% of platinum metals (not just RuO₂) obtained from a melter floor sample. Their micrographs of glass samples exhibited shapes not dissimilar to the "hairball" shapes in RSM and ESM glass samples. The corresponding volume fraction for their data cannot be determined accurately from the data presented in the paper, but it should be about 5 vol%. Alternately, ESM floor samples showed a maximum of about 6 wt% total noble metals and just over 5 wt% maximum for RuO₂. This latter value can be converted to $5 \text{ wt}\% = (0.05 \text{ kg-RuO}_2/\text{kg-glass}) * (2170 \text{ kg-glass/m}^3)/(6970 \text{ kg-RuO}_2/\text{m}^3) = 1.5 \text{ vol}\%$ (based on density numbers consistent with the HWVP melter model computer analysis). Note that in Eq. (1), the sludge layer depth is inversely proportional to $C_{v,max}$. If the ESM data point (1.5 vol%) was assumed to be representative of the maximum packing factor and used in Eq. (1), the sludge layer depth as a function of time shown in Figure 2 is a factor of $(0.06/0.015 = 4.0)$ times greater than shown. This would translate directly to a decrease in the estimated time to reach a postulated failure by the same factor of 4.

Material properties, especially electric conductivity, affect the electrical characteristics upon which estimated times-to-failure are based. The electrical conductivity used in the present work is based on measured data. Other measured electric conductivity data show variations by as much as a factor of 2 from that used in the present work. The variation is in a direction that would tend to indicate that the estimated time to failure given above may be under estimated by a corresponding factor of 2.

In the computer model, an electrode spacing of 127 cm was used. This compares to the HWVP melter electrode spacing of 142 cm. This difference results in the computer model having a smaller volume and smaller floor area (1.669 m^2). Relative to mass accumulation on the floor, these are offsetting effects and the rate of increase of sludge layer thickness would be inversely proportional to floor area as shown by Eq. (1). Thus, the time required for the sludge layer thickness to reach a given depth would be decreased proportionally. This factor, however, is small relative to the variability estimates discussed above.

The predicted melter lifetimes reported in this letter are very short. It is very difficult to qualify these results with existing experimental data. As a rough approximation we can compare it with information from the IDMS melter at Savannah River. As of April, 1993, the IDMS melter had processed 24.5 kg of noble metals with very little electrical disturbance. The IDMS melter has a glass surface area of 0.29 m^2 or 11% that of the HWVP melter. Using the surface area as a scale-up factor the noble metals processed in the IDMS melter would equate to 223 kg in the HWVP melter. The amount of noble metals estimated to cause failure in 162 days of operation in the HWVP melter would be $162 \text{ days}(0.166 \text{ kg total NM/hr})(24 \text{ hr/day}) = 645 \text{ kg}$. This indicates that IDMS melter would have to feed almost three times the current amount of noble metals to fail based on the scenarios mentioned above. It is not clear how long the IDMS melter would have to operate to see an electrical disturbance. This is highly dependent on melter geometry and the form of the settled noble metals layer.

As a similar comparison, the Engineering-Scale Melter (ESM) operated for 60 days in 1992. The ESM has surface area of 0.28 m^2 (11% scale) and processed 9.7 kg of noble metals. At the end of the run a reduction in the resistance between the lower set of electrodes was noticed indicating that a short circuit was forming. 9.7 kg of noble metals in the ESM equates to only 88 kg of total noble metals in the HWVP melter or 22 days of continuous operation (32 calendar days).

The MPA will continue in FY 1994. This will include the completion of the TEMPEST modeling and a complimentary engineering study. The results of the modeling, the engineering analysis, and the GFT, RSM, and ESM experimental studies will be summarized in the preliminary MPA report and the final MPA report. These reports will contain a prediction of melter life based on a combination of the above mentioned information. The information contained in this letter is only the best estimate of melter lifetime at this point in time without any consideration given to the engineering analysis or supporting information.

Figure 1. ESM RuO₂ Material Balance. Results are based on the use of unverified software. No assurance is expressed or implied as to the accuracy, completeness, or usefulness of this information.

Figure 2. HWVP Melter RuO₂ Material Balance. Results are based on the use of unverified software. No assurance is expressed or implied as to the accuracy, completeness, or usefulness of this information.

Figure 3. HWVP Melter Sludge Layer Buildup. Results are based on the use of unverified software. No assurance is expressed or implied as to the accuracy, completeness, or usefulness of this information.

Figure 4. HWVP Melter Operational Parameters - Temperature, Power, and RMS Current. Results are based on the use of unverified software. No assurance is expressed or implied as to the accuracy, completeness, or usefulness of this information.

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