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VOLUME CALIBRATION***

by

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FUEL CONDITIONING FACILITY ELECTROREFINER VOLUME CALIBRATION

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ABSTRACT

In one of the electrometallurgical process steps of the Fuel Conditioning Facility (FCF), the in-process nuclear material is dissolved in the electrorefiner tank in an upper layer of a mixture of liquid LiCl-KCl salt and a lower layer of liquid cadmium. The electrorefiner tank, as most process tanks, is not a smooth right-circular cylinder for which a single linear volume calibration curve could be fitted over the whole height of the tank. Rather, the tank contains many internal components, which cause systematic deviations from a single linear function. The nominal operating temperature of the electrorefiner is 500°C although the salt and cadmium are introduced at 410°C.⁽¹⁾ The operating materials and temperatures preclude multiple calibration runs at operating conditions. In order to maximize the calibration information, multiple calibration runs were performed with water at room temperature. These data allow identification of calibration segments, and preliminary estimation of the calibration function and calibration uncertainties. The final calibration function is based on a combination of data from the water calibrations and the measurements made during the filling of the electrorefiner with salt and cadmium for operation.

INTRODUCTION

Material control and accountancy of special nuclear material in a nuclear facility, with process steps involving liquids, requires an accurate measurement of the liquid content of the process tanks. In the electrometallurgical refining process step of the Fuel Conditioning Facility (FCF), the in-process nuclear material is dissolved in the electrorefiner in an upper layer of liquid salt and a lower layer of liquid cadmium. Thus, to estimate the in-process inventory of special nuclear material in the electrorefiner requires the estimation of an equation, which relates the tank's measurement system, such as in the case of the electrorefiner the liquid level probe, to the volume of the liquid.

The electrorefiner tank, as most process tanks, is not a smooth right-circular cylinder for which a single linear volume calibration curve can be fitted over the whole height of the tank. Rather, the tank contains many internal components, as shown in Fig. 1, which cause systematic deviations from a single linear function. Thus, an important objective of the tank volume calibration is to identify those tank levels over which a single, preferably linear, function will suffice to define the volume versus height relation. The adverse consequences of the perturbations, due to tank internal structures, are expected to play an important role in the case of the electrorefiner. For one, the operating volume of the electrorefiner is generally smaller, by well over an order of magnitude, when compared, for example, to the volume of process or accountancy tanks in aqueous processes. The small size of the tank, therefore, will accentuate the contribution of structural imperfections to the estimated uncertainties. In addition, operational considerations constrain the size and number of incremental additions of liquid to the electrorefiner, and thereby, limit the resolution.

Fig. 1. Electrorefiner Internals

The salt and cadmium, the operating liquids of the electrorefiner, are loaded into the electrorefiner in the argon cell in solid form. They are liquid at the operating temperature of about 500°C. This situation precludes multiple calibration runs at operating conditions. As a matter of fact, the measurements taken during the loading constitute the calibration data. Furthermore, because of the possibility of severe asymmetric thermal stresses on the tank during loading, the loading takes place at a temperature of 410°C, which is about 100°C below the operating temperature of the electrorefiner.

In order to overcome some of these short-comings, two calibrations were performed -- one, consisting of three runs, performed out-of-cell with water at room temperature, the other, consisting of one run, was performed in-cell with the operating fluids at 410°C. Although these two calibrations have some operational differences, we believe they are both, not only relevant, but necessary to the development of a robust measurement function for material accountancy in the Fuel Conditioning Facility.

ANALYSIS OF THE WATER DATA AT ROOM TEMPERATURE

Before the electrorefiner was installed in the argon cell of the Fuel Conditioning Facility, where all operations with the electrorefiner must be performed remotely, the electrorefiner volume versus liquid level relationship was studied using water. The ease of access, and room temperature conditions, in the out-of-cell environment allowed for multiple runs and more incremental additions of liquid than would be practical under in-cell conditions. Three calibration runs, consisting of a series of incremental additions of known masses of water, were made. The basic data -- incremental mass of water, liquid level, and liquid temperature -- form the basis of the calibration. Prior to each run, the electrorefiner was completely emptied.

Three runs of the water calibration measurements were made. For runs one and two, 30 liquid level measurements were made of the first incremental addition and again of the last incremental addition. For each of the 30 incremental additions of liquid in between, only three liquid level measurements were taken. In these two runs the volume increments were about 13 liters each, which resulted in a final volume of about 416 liters and a liquid level of 21 in. The 30 measurements of the same liquid

level at the beginning and end of these two runs served to estimate the variance in the level probe measurements. The third run was designed to give a finer resolution of the height versus volume relation over the first 7.1 in. and the last 3.2 in., by adding the water in increments one fourth of those in runs one and two. In between, the volume increments of water were the same as in the previous two runs.

DATA STANDARDIZATION

The data in the three water calibration runs were standardized to a temperature $T_0 = 20^\circ\text{C}$, and are shown in Figs. 2 through 4.

Fig. 2. Standardized Values -- Run 1

Fig. 3. Standardized Values -- Run 2

Fig. 4. Standardized Values -- Run 3

During the calibration runs only the temperature of the liquid was measured. For data standardization this temperature was assumed to be also applicable to the liquid level probe length and the tank volume expansion. The maximum deviation of the temperature of the measurements from the reference temperature is 4.6°C. This results in an adjustment which is less than the readability of the liquid level gauge. It must be kept in mind, however, that the electrorefiner tank volume calibration with the eutectic mixture LiCl-KCl will take place at 410°C and the operation of the electrorefiner will be at 500°C. Under these circumstances the adjustments will be significant.

DATA SCREENING

The volume $V(h_0)$ of the tank below some level h_0 is given, in general, by the expression

$$V(h_0) = \int_0^{h_0} A(h)dh, \quad (1)$$

where $A(h)$ is the cross sectional area of the tank at level h . If $A(h)$ is constant, the volume of the tank is linear in h .

The engineering design drawing of the electrorefiner tank (see Fig. 1) indicates, significant structures inside the tank, which will result in a nonuniform cross sectional area of the tank with elevation. From Fig. 1 we would expect discontinuities in the tank cross section at least at the 2, 6-3/4, 8-1/2, 15-3/4, 17-1/2 and 20 inch levels; also, a gradual change in the cross sectional area occurs around 5 inches due to the conical surface of the bottom shaper.

A measure of the predictive power of the data, and the eventual correlations derived from it, is its ability to identify the points of discontinuity in the cross sectional area of the tank. To this end, two types of diagnostic plots are very helpful. The first, shown in Fig. 5, and referred to as a profile plot, is a plot of the residuals from a linear fit to all the calibration data versus the liquid level probe measurement.

Fig. 5. Regression Analysis on All Runs

The tank heights in Fig. 5, which were previously identified with changes in tank cross sectional area from the engineering drawings, are clearly supported by the profile plots. However, the profile plots strongly indicate an additional level at ~5 in. at which the tank's cross sectional area changes. This region lies in Fig. 1 approximately between the 4.5 in. and 5.0 in. levels.

We also note that the data for the three runs do not coincide. That is, the between-run differences are significantly greater than the within-run measurement errors. A method has been prescribed for the statistical alignment of data from several runs.⁽²⁾ The procedure is based on choosing an initial segment of the data, which appears to be linearly related, and fitting a single linear function to the data from all runs. The volume or height adjustment for each run is then the ratio of the mean of the residuals for the run to the slope of the regression or merely the mean of the residuals, respectively. The result of this procedure is shown in Fig. 6. We note that above a height of 16 in. the data are still not fully aligned. This would suggest that the systematic behavior, which underlies the nonalignment below 16 in., is not the same as the one above 16 in. However, calculations based on different initial segments of data have shown that residual nonalignment, as the one above 16 in. in Fig. 6, is sensitive

to the choice of data for the linear fit. Some residual nonalignment, although not at the same heights, always appears to remain for these data.

Fig. 6. Calibration Alignment for All Runs

The second diagnostic plot, shown in Fig. 7, is based on the fact that, if we differentiate the expression in Eq. (1) with respect to height, the derivative is equal to the cross sectional area. The incremental slope for the data in the three calibration runs is shown in Fig. 7 as a function of liquid level. As in the previous two plots, the levels at which we should expect the changes in the tank cross sectional area, based on the engineering drawing, are also indicated. Again, these can be identified with changes in the cross sectional area. In addition, the incremental slope plot confirms the previous observation that there is a significant change in the tank cross sectional area a little below the five inch level.

Fig. 7. All Run Incremental Slope

ESTIMATION OF THE CALIBRATION FUNCTION AND CALIBRATION UNCERTAINTY

Although during calibration the volume of liquid added is the controlled variable and the liquid height the measured quantity, we shall follow common practice⁽³⁾ and use the following model

$$V_i = \alpha + \beta h_i + \epsilon_i, \quad (2)$$

where V_i is the volume after the i -th incremental volume addition, h_i the liquid level measurement, and ϵ_i the error. This form of the calibration function is also the measurement function, and consequently inversion is not necessary.

A variety of statistical error models and fitting methods are found throughout the literature.^(3,4) Since the tank is filled in incremental additions, any errors associated with previous additions of liquid can, in principle, affect the error in the total amount. In such cases, for example, the error model chosen for the calibration is a cumulative model.⁽⁵⁾ The cumulative error model appears to be the most frequently chosen model, both for statistical reasons and computational implications. Based on the analysis of different possible models and the fact that at the high operating temperature of the electrorefiner measurement error is sure to be larger, we choose Ordinary Least Squares as the estimation method.

The estimated calibration functions, based on run three data, together with the estimates of the random and systematic error variances for the linear segments, are shown in Table I.

ELECTROREFINER VOLUME CALIBRATION BASED ON SALT AND CADMIUM AT 410°C

We now compare the data collected during the loading of the electrorefiner with the working fluids -- salt and cadmium, and the associated calibration estimates, to the results based on measurements with water at room temperature. Since loading the electrorefiner with salt and cadmium precludes multiple runs, the three calibration runs with water at room temperature as the working fluid, are an important compliment to the inferences based on salt and cadmium data. As such, every effort is made to incorporate the information in the water data into the final salt and cadmium calibration and measurement functions.

Table I. Estimated Calibration/Measurement Function Using Ordinary Least Squares Model for Out-of-Cell Volume Calibration Run #3 (Reference Temperature 20°C)

Liquid Height, H (in.)	Number Data Points	Calibrated Volume, V (ℓ)	Random Variance, σ_R^2 (ℓ ²)	Systematic Variance, σ_R^2 (ℓ ²)
$2.00 \leq H < 4.62$	14	$-1.96280 + 17.9464 \times H$	8.31013×10^{-4}	$6.41259 \times 10^{-2} - 2.87980 \times 10^{-2} \times H + 6.85471 \times 10^{-3} \times H^2$
$4.62 \leq H < 6.75$	14	$-11.3153 + 19.9846 \times H$	1.03049×10^{-3}	$5.15948 \times 10^{-1} - 1.53021 \times 10^{-1} \times H + 1.65253 \times 10^{-2} \times H^2$
$8.5 \leq H < 15.75$	11	$-13.9485 + 20.3193 \times H$	1.06530×10^{-3}	$1.66336 \times 10^{-1} - 2.15882 \times 10^{-2} \times H + 1.19884 \times 10^{-3} \times H^2$
$17.5 \leq H < 21.25$	21	$-15.6185 + 20.3905 \times H$	1.06903×10^{-3}	$1.60881 - 1.51891 \times 10^{-1} \times H + 4.22053 \times 10^{-3} \times H^2$

Comparison to the Water Data

In Fig. 8 we shown the salt-cadmium residuals, relative to the regression curve based on the water data, over the full height of the salt-cadmium volume in the electrorefiner. It is clear that a measurement function based on the salt-cadmium data will systematically predict a greater liquid volume than one based on the water calibration data. Moreover, when viewed from a slightly different perspective as in Fig.9, where the salt-cadmium measurements and the water measurements are subtracted from a common arbitrary regression line over the height of the electrorefiner, the difference between the two data increases with liquid level. The rate of increase is greater for the cadmium additions than for the salt additions. The systematic nature of this difference suggests that there may be a dominant component.

It has been noted by the operators, that during the initial incremental additions of the cadmium to the electrorefiner, there was some "splashout" of salt. Furthermore, there occurred some spillage of cadmium during the removal of the bulk loader after the second increment (i.e., for the third increment). We quantify and bound these possibilities in the light of the measurements. To this end, Fig. 10 demonstrates, for the salt level measurements, the effect of including the maximum

estimate for each anomaly -- a splashout of 1 kg of salt at each of the first two cadmium incremental additions (1.21 total), and a spillage of 0.51 of cadmium after the second increment. These adjustments remove the slope discrepancy for the first five of the nine cadmium increments. The spillage of 0.51 of cadmium also removes most of the bias for the ascending cadmium level data.

Fig. 8. Volume Residuals, Relative to Water Calibration Regression Line, for Salt Level (o) Measurements for all Salt and Cadmium Increments and for Water Level (•) Measurements of Water Calibration

Fig. 9. Volume Residuals, Relative to Arbitrary "Calibration" ($V=0+20 \cdot H$) Line, for Salt Level (o) Measurements for all Salt and Cadmium increments and for Water Level (•) Measurements of Water Calibration

Fig. 10. Volume Residuals, Relative to Water Calibration Regression Line, for Salt Level (o) Measurements for all Salt and Cadmium Increments Assuming Maximum Estimates for Observed Splash/Spill Losses (Salt Loss of 0.6 l at each Cadmium Increment #1 and #1 and Cadmium Loss of 0.5 l at Cadmium Increment #3)

Estimation of the Measurement Function and the Measurement Uncertainty

Of interest to material accountancy in the electrolyzer are the cadmium volume and the salt volume. These are determined, via measurement functions, by the two liquid levels -- the top of the cadmium and the top of the salt. The current operating levels are a minimum, and are expected to slowly increase as operation proceeds until drawdown of the electrolyzer. It is, therefore, sufficient to focus the estimates on the expected ranges of the liquid levels over that period.

Top of the Salt

The estimate is based on liquid level measurements in the range 13.3 in. to 16.4 in. We estimate that in the range of liquid levels from 13.3 in. to 17.5 in. the following measurement function is appropriate:

$$V_{sc} = 21.9016 + 21.2383 \cdot h_{ts} \quad (3)$$

where V_{sc} is the salt and cadmium volume in liters and h_{ts} the top of salt liquid level in inches. The associated estimated random error for the volume estimate in liters is $\sigma_r = 0.214$. The estimate of the systematic error in liters is given by

$$\sigma_s = \{1.80 - 0.231 \cdot h_s + 0.00746 \cdot h_{ts}^2\}^{1/2} \quad (4)$$

Top of the Cadmium

The estimate is based on liquid level measurements in the range 2.0 in. to 4.0 in. We estimate that in the range 2.0 in. to 4.65 in. the following measurement function for the cadmium volume V_c in liters is appropriate:

$$V_c = 0.3828 + 17.4334 \cdot h_{tc}, \quad (5)$$

where h_{tc} is the cadmium liquid level in inches.

The associated estimated random error for the volume estimate in liters is $\sigma_r = 0.335$. The estimate of the systematic error in liters is given by

$$\sigma_s = \{0.698 - 0.411 \cdot h_{tc} + 0.0619 \cdot h_{tc}^2\}^{1/2}. \quad (6)$$

Volume of Salt

The volume of salt between the levels h_{ts} and h_{tc} is given by the difference of Eqs. (4) and (5),

$$V_s = V_{sc} - V_c = 21.5188 + 21.2383 \cdot h_{ts} - 17.4334 \cdot h_{tc}. \quad (7)$$

The associated random error is $\sigma_r = 0.397$ liters. The systematic error is given by

$$\sigma_s = \{2.498 - 0.411 \cdot h_{tc} - 0.231 h_{ts} + 0.0619 \cdot h_{tc}^2 + 0.00746 \cdot h_{ts}^2\}^{1/2}. \quad (8)$$

Volume of Cadmium

The volume of the cadmium is determined by the relations given above as Eq. (5) and the associated random and systematic errors.

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