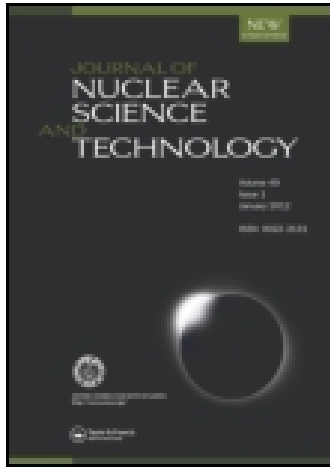


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TECHNICAL REPORT

Long Term Change in Level-Volume Relation of an Accountability Tank for Plutonium Nitrate Solution

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A slight change in the level-volume relation for an accountability tank for a large amount of plutonium nitrate solution (PuN) was observed at the Plutonium Conversion Development Facility (PCDF) in the Power Reactor and Nuclear Fuel Development Corp. (PNC), Tokai Works. From the results of annual tank re-calibrations for the plutonium receiving tank from 1985 to 1992 using the incremental feed of nitric acid as the density standard, it became clear that the relation between the level and the volume changed slightly, and the rate of the change was a linear function of operating time. Also it became clear that the change was linear in relation to the level. In the PCDF, the cumulative change in the volume at the nominal level was evaluated to be 0.1% during 8 years' operation. It was also evaluated that the repeatability of the re-calibration is much better than 0.1%. A reasonable frequency of tank re-calibration is once every 5 years.

KEYWORDS: *volume measurement, accuracy, long-term change, plutonium nitrates, accountability tank calibration, manometer, safeguards, reprocessing, plutonium conversion, nuclear fuel conversion*

I. INTRODUCTION

It is known that the error in volume measurement at the input or the output accountability tank in a nuclear reprocessing and Pu conversion plant is a major part of the total accounting error. According to the international standards of accountancy described in the safeguards criteria (1991~1995) of the International Atomic Energy Agency (IAEA), the expected measurement accuracy associated with closing a material balance in a Pu conversion plant is 0.5% including volume measurement of the plutonium nitrate solution (PuN), weight measurement of the powder, and sampling and analysis of the solution and the powder. Thus, the target value for the systematic error of the volume measurement is set to be 0.2%. Moreover, this value must be main-

tained throughout plant operation by periodic calibration of the tank and the level measurement equipment. The tank re-calibration is to check the change in the relation between the level and the volume. The relation is expressed as a polynomial equation, known as the volume measurement equation. There is a possibility of the relation changing because of the corrosion of the inner surface of the tank or the slight deformations of the tank. Tank re-calibration, therefore, is an important inspection matter for the IAEA.

However, tank re-calibration yields a lot of liquid waste and greatly interferes with process operation. From the viewpoint of process operation, it is desirable that the frequency of re-calibration is as low as possible. Thus, it is very important to determine the rate of the change in the relation and to

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decide the reasonable frequency of tank re-calibration. In this paper, these values are reported for 8 years' operation at the Plutonium Conversion Development Facility (PCDF) in the Power Reactor and Nuclear Fuel Development Corp. (PNC), Tokai Works.

II. OUTLINE OF THE PCDF

1. Process

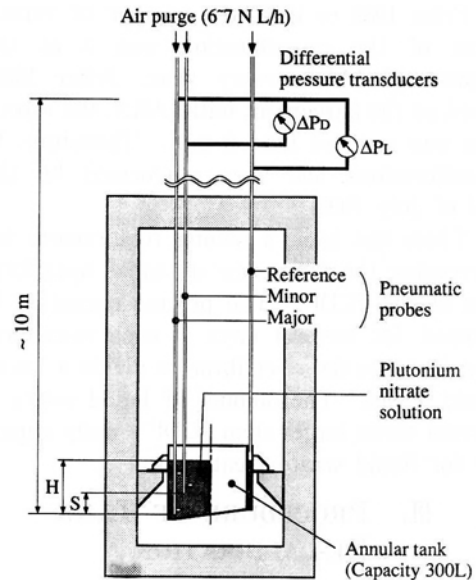
The PCDF is the first co-conversion demonstration plant in the world using the microwave heating method⁽¹⁾. The process is very simple and very little liquid waste is obtained. The actual throughput of the PCDF is 600~700 kg of Pu per year. The batch size of PuN transferred from the Tokai Reprocessing Plant (TRP) is 50~60 kg of Pu. The TRP and the PCDF are connected by pipelines and the amount of PuN transferred from TRP to PCDF is determined at the input accountability tank in the PCDF. Therefore, there is no shipper/receiver difference (S/RD) between these facilities. The PCDF started operation in 1984 and the cumulative throughput of Pu was over 3,700 kg at the end of March 1993.

There are four annular tanks for PuN in the PCDF. One is the accountability tank and the others are the PuN buffer tank, the PuN-uranyl mixing tank and the PuN-uranyl buffer tank. All have the same shape with a nominal liquid volume of 300 l and a nominal liquid level of 1,200 mm.

2. Tank Calibration and Volume Measurement System

Figure 1 shows the volume measurement system. The level and the density of solution in the tank are measured by air sparging and differential pressure measurement. The volume measurement equation (F) necessary for converting level to volume and the separation length (S) are determined during the initial calibration before starting operation by air sparging and differential pressure measurement using an incremental feed of pure water as the density standard. The volume measurement equation and the separation length are checked during re-calibration using an incremental feed of nitric acid with a known

density as the density standard and, if necessary, the equation and the separation length are revised. This method is frequently used in many nuclear reprocessing and Pu conversion plants⁽²⁾.



$$\Delta P_D/k = \rho \times g \times S, \quad \Delta P_L/k = \rho \times g \times H, \quad V = F(H)$$

ΔP_D : Differential pressure (density)
 ΔP_L : Differential pressure (level)
 ρ : Liquid density, V : Liquid volume
 H : Level height, S : Separation length
 g : Local gravitational acceleration (979.963 m/s²)
 k : Constant (0.9985)
 F : Volume equation
 (3rd order polynomial equation)

Fig. 1 Volume measurement system

The basis of volume measurement is an accurate differential pressure measurement. From 1982 to 1984, the authors have developed an accurate pressure measurement system using a pair of differential digital quartz pressure transducers⁽³⁾⁽⁴⁾. The pressure measurement error can always be less than 2 Pa including random and systematic errors with a periodic calibration once a year. The accuracy of the transducers is checked by an air-piston pressure gauge at every volume measurement and tank calibration.

In the PCDF, the initial calibration was done in 1982 for all the PuN tanks to determine the volume equation and the separation length. Re-calibration has been done for only the input accountability tank since 1985, and

the volume equation and the separation length have never been revised, because the level-volume relation checked by re-calibration was in good agreement with the volume equation determined at initial calibration.

From 1985 to 1988, the number of repetitions of the re-calibration was 3 at the request of IAEA every year. After 1989, based on the agreement with IAEA, the repetition was reduced from 3 to 1. Therefore, 16 re-calibrations had been performed by the end of July 1992.

There has been a strong requirement for decreasing the frequency of tank re-calibration at the PCDF. The process operation is stopped for several days at each re-calibration, because the re-calibration yields a lot of liquid waste. The amount of liquid waste is several times larger than PCDF's daily capacity for liquid waste treatment.

III. PROCEDURE OF TANK RE-CALIBRATION

1. Tank Re-calibration

Figure 2 shows the procedure for checking the volume equation. Figure 3 shows the procedure for checking the separation length. To check the volume equation, the difference between "Volume 1" and "Volume 2" at each level is evaluated. "Volume 1" is determined by the cumulative weight of nitric acid and the density of nitric acid corrected for the temperature inside the tank. "Volume 2" is determined by the pressure measurement, the corrected density of nitric acid and the volume equation. The heel volume at the

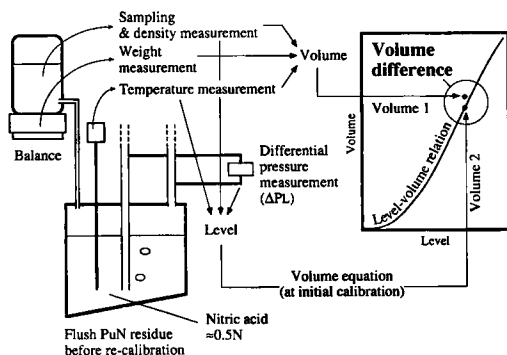


Fig. 2 Procedure to confirm volume equation

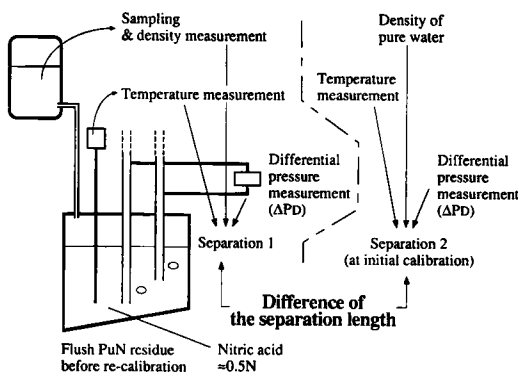


Fig. 3 Procedure to confirm separation length

bottom of the tank before re-calibration is added to "Volume 1". A long term change in level-volume relation was revealed from a detailed analysis of the volume differences.

To perform a precise tank re-calibration, it is very important to flush the PuN residue inside the tank sufficiently before re-calibration. The PuN residue on the slightly sloping bottom of the tank or on the wall introduces uncertainty in the density of nitric acid which is fed to the tank as the density standard. The authors estimated the amount of PuN residue on the bottom and the wall based on operation data and washed the tank twice with minimum waste liquid. The amount of waste liquid is about 70 l for the wash.

2. Equipment

(1) Accountability Tank

The nominal volume of the PCDF's annular shaped accountability tank is 300 l for a liquid level of 1,200 mm. The outer diameter of the tank is 1,600 mm, the thickness of PuN is 59 mm and the height of the tank is 1,400 mm. The material is 8 mm thick low carbon stainless steel with a passivated surface finish. The inner diameter of the dip-tube is 12 mm and the dip-tube is supported very firmly from the wall of the tank. The air flow rate for differential pressure measurement is 6~7 Nl/h, and the bubbling frequency is about twice every 3 s. The nominal separation length for density measurement is 200 mm.

For PuN with a density about 1.5 g/cm³, the differential pressure for the density measurement (ΔP_D) is about 3,000 Pa and the dif-

ferential pressure for the level measurement (ΔP_L) is about 17,000 Pa at the nominal level. These transducers are set about 10 m above the level of tank, so the observed differential pressure is a little smaller than the true differential pressure, because the air-density is distributed along the dip tube. The local gravitational acceleration is 979.963 m/s².

(2) Balance

A load-cell type balance is used. Manufacturer's specifications are shown in **Table 1**. To measure the weight of nitric acid correctly, the top-plate of the balance is reinforced using a thick steel plate. Buoyancy corrections are made for both the standard weights and the nitric acid. The balance is calibrated from 0 to 100 kg using high-grade standard weights at 20 kg intervals. The span adjustment is done for each re-calibration.

Table 1 Specification of the balance

| | |
|---------------|-------------------|
| Full scale | 0~100 kg |
| Resolution | 0.01 kg |
| Repeatability | 0.01% full scale |
| Hysteresis | ±0.01% full scale |
| Non-linearity | ±0.01% full scale |

(3) Differential Pressure Transducer

Digital quartz differential pressure transducers are used. Manufacturer's specifications are shown in **Table 2**. To obtain an accuracy comparable to the conventional quartz bourdon transducer by Ruska⁽⁵⁾⁽⁶⁾, the authors have optimized the various settings of the transducer, such as optimization of the non-linearity within the measurement range, temperature stabilization and auto zero-adjustment, and relaxation of the pressure oscillation. For each PuN receipt and tank calibration or re-calibration, the transducers are calibrated from 0 to 20,000 Pa at 5,000 Pa intervals, using

Table 2 Specification of the pressure transducer

| | |
|---------------------------------------|-----------------------|
| Pressure range for ΔP_L | 0~15 psi differential |
| Pressure range for ΔP_D | 0~6 psi differential |
| Repeatability | 0.005% full scale |
| Hysteresis | 0.005% full scale |
| Temperature null shift | 0.0007% full scale/°C |
| Temperature sensitivity shift | 0.0049% reading/°C |
| Common mode error | 0.002% full scale/psi |
| Common mode pressure (maximum) | 900 psi |
| Frequency excursion (0 to full scale) | 40~36 kHz |

the air-piston pressure gauge authenticated by the Japanese National Research Laboratory of Metrology (NRLM). The frequency of span adjustment is once a year.

(4) Nitric Acid and Density Meter

0.5N nitric acid is used as the density standard. To minimize the density variation, the solution is stored overnight in a room where the room temperature is precisely controlled. To measure the density, a vibration type density meter has been used since 1989. Manufacturer's specifications are shown in **Table 3**. Before 1988, the density was calculated by acidity and temperature measurement.

Table 3 Specification of the density meter

| | |
|---------------------------|---------------------------------------|
| Full scale | 0~3 g/cm ³ |
| Error | ±1×10 ⁻⁵ g/cm ³ |
| Temperature control error | ±0.02°C |

(5) Thermometer Inside Tank

A thermocouple covered with a stainless steel tube is installed inside the tank. The measurement error including the transmitter is 0.5°C. The transmitter is calibrated once a year.

3. Analytical Procedure

(1) Errors in Measurement Equipment

Based on the records of tank re-calibrations from 1985 to 1992, verified by IAEA inspectors, random and systematic errors in the balance and the pressure transducers (ΔP_L and ΔP_D) were calculated.

(2) Error Composition

The difference between "Volume 1" and "Volume 2" is a function of the level. **Figure 4** is a conceptual sketch of the difference.

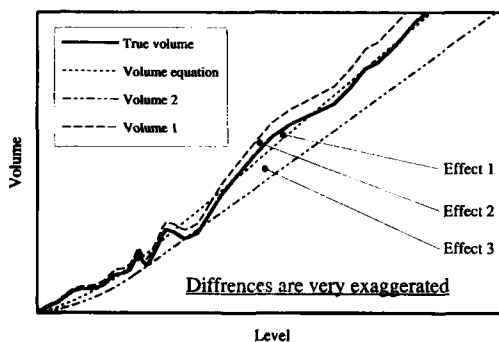


Fig. 4 Conceptual figure of differences between Volumes 1 and 2

The difference includes three components ;

- Effect 1: Difference between the true volume and the volume equation, including the fitting error of the equation and change in the volume equation.
- Effect 2: Difference between the true volume and "Volume 1", including measurement errors for weight and density of nitric acid.
- Effect 3: Difference between the volume equation and "Volume 2", including measurement errors for pressure and density of nitric acid.

The effect of tank thermal expansion is negligible, because the temperature difference between the initial calibration and the re-calibration is less than 4°C. In addition to these errors, there could be the effect of losses such as leakage of nitric acid.

(3) Regression Analysis

Figure 5 shows an example of the result of volume difference plotted as a function of the level. The difference can be analyzed by simple linear regression. The gradient of the slope indicates the difference in the level-volume relation between the initial calibration and the re-calibration. The gradient of the slope also includes the systematic measurement error of Effect 2 and 3. The deviation from the slope indicates the fitting error and the random measurement error of Effect 2 and 3, because they have no correlation with the level. The change in the gradient of the slope as a function of the elapsed time means

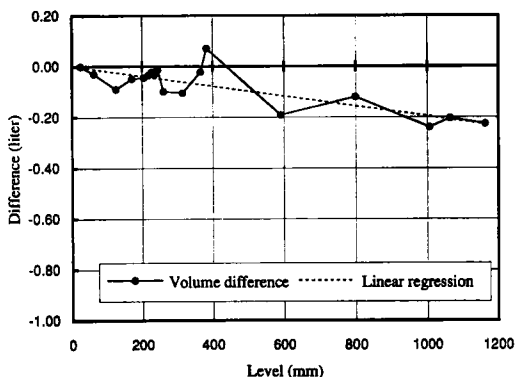


Fig. 5 Example of volume difference

a long term change in the level-volume relation.

IV. RESULTS

1. Errors in Measurement Equipments

The random and systematic errors for each item of equipment are shown in Table 4. All equipment has been kept in good condition because the systematic error is smaller than the random error. For the balance, the systematic error is 0.013% of full scale, and the standard deviation of the random error is 0.012% of full scale. For the level pressure transducer, the systematic error is about 0.5 Pa, that is 0.004% of the nominal pressure (~12,000 Pa) for tank re-calibration or 0.003% of the nominal pressure (~17,000 Pa) for PuN measurement. For the density pressure transducer, the systematic error is 0.6 Pa, that is 0.03% of the nominal pressure (~2,000 Pa) for tank re-calibration or 0.02% of the nominal pressure (~3,000 Pa) for PuN measurement. For both the pressure transducers, the standard deviation of the random error is 1 Pa.

Table 4 Summary of instrument calibrations from 1985 to 1992

| Instrument | Random error (1σ) | Systematic error |
|------------------------------|----------------------------|------------------|
| Balance | 0.012%FS | 0.013%FS |
| Digi-quartz (ΔP_L) | 1.2 Pa | 0.5 Pa |
| Digi-quartz (ΔP_D) | 0.8 Pa | 0.6 Pa |

2. Volume Difference

Figure 5 shows the volume difference between "Volume 1" and "Volume 2" in 1992 plotted as a function of the level. The difference is defined as Volume 2 minus Volume 1. Thus, Fig. 5 shows that Volume 1 is larger than Volume 2. Since 1985, the volume difference has been very similar to Fig. 5, and a step change in the difference caused by liquid loss has never been observed. It seems that the volume difference increases linearly with level, so the linear regression was applied.

3. Fitting Error and Random Measurement Error

Figure 6 shows the annual accumulation of differences between measurements and the regression line from 1985 to 1992. The solid

curve indicates a fitting error in the volume equation. The distribution around the solid curve indicates random errors in weight and pressure measurements. The maximum and minimum values of the fitting error are 0.15 l and -0.08 l. At the nominal level, the fitting error is negligible. The random error decreases with the level. The standard deviation in the random error is less than 0.1 l, which is smaller than the fitting error. At the nominal level, the random error is 0.04 l (0.013% for the nominal volume), which is nearly equal to the random error in the measurement equipment.

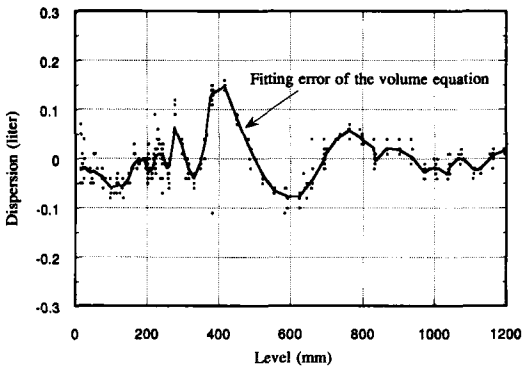


Fig. 6 Accumulation of differences from regression line

4. Change in Level-volume Relation and Systematic Measurement Error

Figure 7 shows the change in the gradient of the linear regression as a function of elapsed operating time. The number of PuN transfers from the TRP to the PCDF was

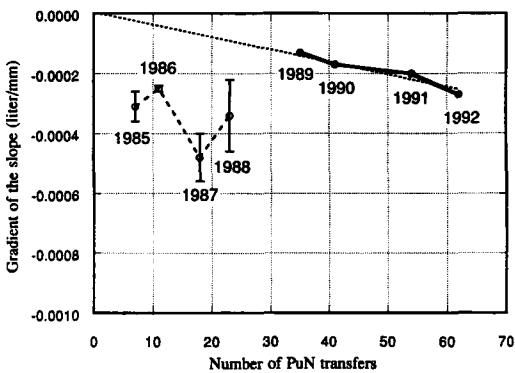


Fig. 7 Change in gradient of linear regression line

used as the elapsed operating time, because the corrosion or the deformation of the tank may be related to the number of transfers.

From 1985 to 1988, the re-calibration was repeated 3 times and the error bar indicates the standard deviation for the 3 runs. In 1989, the 3 run calibration was discontinued and replaced by the precise density meter. The systematic error limit for the volume difference due to the measurement equipment was evaluated to be 0.1% for the nominal volume, that is 0.00025 l/mm in terms of gradient of the slope. From 1989 to 1992, the gradient was within the systematic error limit every year.

Moreover, it is clear that the gradient of the slope has changed very slightly and linearly since the start of operation. This means that the thickness of the solution inside the annular tank has become a little larger. Before 1988, the change was not clear because the density measurement error was large and the gradient was very small. The change in the gradient was 0.00024 l/mm during 8 years, which is nearly 0.1% of the nominal volume.

The total systematic error in the tank calibration and volume measurement system is evaluated to be 0.12% from the study of systematic error propagation in the system⁽⁷⁾, excluding the change in the level-volume relation. The change in the level-volume relation is 0.06% during 5 years. Consequently, a frequency of tank re-calibration of once every 5 years is sufficient for keeping the total systematic error within 0.2%.

On the other hand, the distribution around the dotted line is very small, so the repeatability of re-calibration was evaluated to be much better than 0.1%.

5. Separation Length

After 1989, the maximum of the difference between "Separation 1" and "Separation 2" has been 0.25 mm (0.12% of the initial length). A yearly change has never been observed.

V. CONSIDERATION

There are two reasons why the volume difference changes linearly with level and also changes linearly with the number of PuN transfers. One is the systematic measurement

error and the other is a real change in the relation between the level and the volume.

If the difference (about 0.1% of the nominal volume) is a result of the systematic measurement error, the major portion of the error is in the density of the nitric acid, because the pressure measurement and weight measurement errors are negligible as shown in Table 4. There is a possibility that the Pu residue left inside the tank before re-calibration dissolves in the nitric acid and increases the density error. In this case, the Pu-concentration in the nitric acid would have to be about 0.6 g Pu/l to give the expected⁽⁸⁾ density change. However, the chemical analysis shows that the concentration is very low.

It is assumed from the results that the change in the relation between the level and the volume is caused by the corrosion of the tank material. The estimated corrosion depth is $\sim 20 \mu\text{m}$, if the material dissolves uniformly. Figure 5 shows that the corrosion depth is constant in relation to the level because the volume difference is linear with level. This corrosion depth is larger than that of the same material immersed in nitric acid under the same conditions in the absence of Pu. This result supports the idea⁽⁹⁾ that Pu accelerates the corrosion rate of stainless steel in nitric acid. In the PCDF, the chemical composition of PuN is $\text{Pu}(\text{NO}_3)_4$, the acidity is approximately 6 N, the concentration of Pu is approximately 210 g/l, the temperature is 30~40°C, the α -activity is approximately 10^{10} Bq/g Pu, and the γ -activity is approximately 10^6 Bq/g Pu. In addition, the inner surface of the tank material is exposed to repeated wet and dry conditions, because the PuN is transferred to the storage tank several days after receipt. The frequency of the receipt and the transfer is about 10 times a year. For a detailed study, additional corrosion tests under such conditions will be necessary.

VI. CONCLUSIONS

Accountability tank re-calibration data and digital quartz pressure transducer re-calibration data were analyzed based on 8 years' field operations from 1985 to 1992 in the PCDF of the PNC Tokai Works.

The systematic error in the digital quartz pressure transducer was about 0.5 Pa, and the random error was about 1 Pa with periodic calibration once a year.

A slight change in the level-volume relation of an accountability tank for a large amount of PuN was observed. The relation between the level and the volume changed slightly and linearly with the number of operations. Also it became clear that the change was linear relative to the level.

In the PCDF, the cumulative change in the volume at the nominal level was evaluated to be 0.1% during 8 years' operation. It was also evaluated that the repeatability of the re-calibration was much better than 0.1%. A reasonable frequency of tank re-calibration is once every 5 years.

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