JAEA-Research
2010-033

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Volume Measurement System for Plutonium Nitrate Solution and its Uncertainty to be Used for Nuclear Materials Accountancy Proved by Demonstration over Fifteen Years

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October 2010
Japan Atomic Energy Agency

日本原子力研究開発機構

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(Received July 29, 2010)

An accurate volume measurement system for plutonium nitrate solution stored in an accountability tank with dip-tubes has been developed and demonstrated over fifteen years at the Plutonium Conversion Development Facility of the Japan Atomic Energy Agency. As a result of calibrations during the demonstration, it was proved that measurement uncertainty practically achieved and maintained was less than 0.1 % (systematic character) and 0.15 % (random) as one sigma which was half of the current target uncertainty admitted internationally. It was also proved that discrepancy between measured density and analytically determined density was less than 0.002 $g \cdot cm^{-3}$ as one sigma. These uncertainties include effects by long term use of the accountability tank where cumulative plutonium throughput is six tons. The system consists of high precision differential pressure transducers and a dead-weight tester, sequentially controlled valves for periodical zero adjustment, dampers to reduce pressure oscillation and a procedure to correct measurement biases. The sequence was also useful to carry out maintenances safely without contamination. Longevity of the transducer was longer than 15 years. Principles and essentials to determine solution volume and weight of plutonium, measurement biases and corrections, accurate pressure measurement system, maintenances and diagnostics, operational experiences, evaluation of measurement uncertainty are described.

Keywords Reprocessing, Conversion, Accountability Tank, Dip-tube, Bubble Pressure, Quartz Pressure Transducer, Electro Manometer, Measurement Bias, Level Measurement, Density Measurement, Safeguards, Solution Monitoring

JAEA-Research 2010-033

硝酸プルトニウム溶液の液量測定システム及び 15年以上の実証運転により証明された計量管理のための測定の不確かさ

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細馬　隆

(2010年7月29日受理)

日本原子力研究開発機構のプルトニウム転換技術開発施設では、浸漬管を持つ計量槽に貯蔵された硝酸プルトニウム溶液の高精度液量測定システムを 15年以上にわたり開発・実証してきた。実証運転中の校正試験の結果から、現実的に達成・維持が可能な測定の不確かさ（1 σ）は 0.1 %（系統的性質）及び 0.15 %（偶然）未満であることを証明した。これは国際的に認知されている現在の値の半分であった。また、測定した溶液密度と分析によって求めた溶液密度の差（1 σ）は 0.002 $g \cdot cm^{-3}$ 未満であることを証明した。これらの不確かさは、計量槽の長期間の使用（累積通過プルトニウム量で 6 t）の影響を含んでいる。測定システムは、高精度の差圧変換器と校正用重錘型圧力計、定期的零点調整のためのシーケンス制御弁、圧力変動減衰のためのダンパー及び測定バイアスを補正するための手順等から構成される。本シーケンスはまた、汚染を発生させずに安全に保守作業を行うためにも有効であった。差圧変換器の寿命は 15年以上と評価された。この資料では、液量及びプルトニウム重量測定の原理及び枢要点、測定バイアスと補正、高精度の圧力測定システム、保守及び診断、運転経験、及び測定の不確かさの評価について述べる。

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1. Introduction

Reprocessing of spent fuels from nuclear power plants and reuse of plutonium to new fuels as mixed oxide (MOX) has been adopted as a national policy in Japan. To proceed practical tests and developments concerning nuclear fuel cycle under an international consensus, it is essential to carry out material accounting very carefully and strictly and to declare all nuclear material under the safeguards by IAEA, which can be sustained by technical developments and efforts to strengthen accountability of plutonium in every chemical or physical form.

Research and development concerning the demonstration of reprocessing process had started in 1977 at the Tokai Reprocessing Plant (TRP) operated by the Japan Atomic Energy Agency (JAEA). The total weight of reprocessed fuel has reached to 1136 metallic tons at the end of 2009(1). Plutonium extracted at TRP is transferred in the form of nitrate solution to the Plutonium Conversion Development Facility (PCDF) adjacent to TRP. The solution is mixed at PCDF with uranyl nitrate solution and co-converted directly to MOX powder by microwave direct denitration method(2)(3)(4). This co-conversion process is the original technology invented and developed in the late 1970s by the Power reactor and Nuclear fuel Corporation the predecessor of JAEA. One of the important advantages of the technology is a significant contribution to nuclear nonproliferation, because the most sensitive material in safeguards, pure plutonium dioxide, is nowhere in the process. Research and development concerning the demonstration of co-conversion process had started in 1983(5). The total weight of plutonium co-converted to MOX has reached to 6.2 tons at the end of 2009. Obtained technical knowledge and operational experiences in both facilities have been applied to the design, construction, consultation and training of personnel of the Rokkasho Reprocessing Plant (RRP) operated by the Japan Nuclear Fuel Limited (JNFL).

There were two major objectives to strengthen the accountability of plutonium at PCDF : 1) Measurement of volume by dip-tube manometry in a tank to receive and/or store plutonium nitrate solution, because its measurement uncertainty was assumed to be the largest source of total measurement uncertainty associated with plutonium accounting. The assumed uncertainty was 0.2 % for systematic character and 0.3 % for random according to the International Target Values (ITVs) for measurement uncertainties in safeguarding nuclear materials(6) proposed by an international working group. These uncertainties were at least twice as large as other measurement uncertainties such as for an electro-balance and for analytical methods. The proposed ITVs were set at the overlapping area of industrial operations (state of practice) and analytical measurements (state of art). ; 2) Direct measurement of plutonium weight by Non-Destructive Assay (NDA)(7) in a waste drum and holdup that is residue of powder in glove boxes, because the weight of plutonium they include had become be recognized to be small but significant to obtain reasonable material balance.

Despite such relatively large measurement uncertainties, the volume measurement performed at the input tank at PCDF is indispensable to determine the weight of plutonium transferred from reprocessing to co-conversion which constitutes the major flow of plutonium recycle. The uncertainties affect seriously the statistical evaluation associated with closing plutonium balance which corresponds to a possibility of unaccounted plutonium that could be suspected of diversion. The evaluation is a comparison test between physical inventory and book inventory calculated from sum of the previous physical inventory and inputs subtracted by outputs afterwards. Its uncertainty is calculated from the sum of squares of each measurement uncertainty, thus over half of the uncertainty were occupied by volume measurement if the assumptions above were most probable. The Japanese national policy to declare all plutonium can only be proved by the consistency of acceptable results of the evaluation by operator, government and IAEA, however the uncertainty inevitably clouds the results. Therefore, it was necessary to reduce the assumed volume measurement uncertainty.

Therefore, an electro-manometer (ELTM) system for the input tank and other storage tanks utilizing a set of high precision digiquartz® pressure transducers independently for each tank had been developed at PCDF since 1982. The purpose was to decrease volume measurement uncertainty to 0.1 - 0.2 % as one sigma which was nearly the same as analytical measurements used for plutonium accounting. The second purpose was to keep always the uncertainty by periodical maintenance as industrial operation. This system has been put into practice and demonstrated in the presence of inspectorate (government and IAEA) since the start of operation. This system had also been used by inspectorate, until installation of their own system, as an inspection device based on a premise that the device is carefully verified using their own standards and/or other approaches. Therefore, the obtained experiences and the results concerning usage, calibration and maintenance have been shared between operator and inspectorate. We have now accumulated enough data of calibrations and other tests to evaluate measurement uncertainty practically achieved and sustained by appropriate maintenance. Improvement of the system is being continued steadily by introducing a new data treatment associated with high temperature solution derived from high burn-up fuel reprocessing. Another ELTM system utilizing a high precision quartz bourdon tube whose output is converted into angular displacements of a small mirror and an improved scanning valve commonly used for several tanks had been developed at TRP as a part of the Tokai Advanced Safeguards Technology Exercise (TASTEX)(8).

For NDA, several devices for waste, holdup and MOX scrap utilizing neutron detectors and a coincidence counter have been developed, verified and validated by inspectorate, put into practice and demonstrated for material accounting collaboratively between JAEA and Los Alamos National Laboratory under DOE since the end of 1980s(9). These devices can measure objects in which representative sample is difficult to be taken, so plutonium weight in every chemical and/or physical form can now be measured. However, most of plutonium weight in the major flow and/or inventory is still determined by destructive analysis, because the best accuracy of the current NDA is considered to be 0.5 - 1.0 %(10) achieved for measurement of clean MOX product. New NDAs are under development to improve accuracy, to extend measurement objects to the major flow and/or inventory, and to achieve more effective and efficient safeguards(11)(12).

The purposes to develop ELTM system were : 1) Achieve volume measurement uncertainty to 0.1 - 0.2 % as one sigma considering some practical bias corrections ; 2) Keep this uncertainty by appropriate maintenances ; 3) Prove this uncertainty as industrial operations by demonstration.

In this report, we describe principles and essentials to determine solution volume and weight of plutonium in a tank, measurement biases and corrections, components of the system and their purposes, maintenances and operational experiences of the system including longevity and diagnostic of the transducer, and evaluation of measurement uncertainty based on accumulated data during the demonstration. Finally, we wish to propose a new set of target values applied to volume measurement uncertainties by dip-tube to be used for nuclear material accountancy, which would supersede the conventional ITVs.

2. Principles and essentials to determine plutonium weight in a tank

Measurement system of volume by dip-tube manometry is a large system consists of an accountability tank and a set of pressure transducers. The tank is conceptually a graduated vessel and the transducers are devices both to graduate the scale corresponds to liquid level and to read the scale. In contrast with many other measurement system to which a superior standard is supplied, the tank can not be compared with such a standard which is effective to decrease measurement uncertainty. The primary standards are the reference pressures used to calibrate the transducers and the reference liquid with known density and weight normally pure water used to calibrate the tank. Thus, propagation of measurement uncertainty becomes very complicated, that is the reason why a large measurement uncertainty has been assumed.

2.1 Accountability tank and equipment

Plutonium nitrate solution purified at TRP is transferred to PCDF through a pipeline, normally in a batch size of 55 kg plutonium. The pipeline is declined toward PCDF and rinsed by nitric acid just after the transfer. Volume of the homogenized mixture of the plutonium nitrate solution and the nitric acid is measured at an annular shaped accountability tank equipped with three dip-tubes (called major, minor and reference in the order of tube length), two pressure transducers to measure ΔP_L and ΔP_D connected differentially to the dip-tubes by conducting pipes, a thermocouple probe, a sampling line and process lines. This tank has no tubes nor lines at the bottom plate and the side wall, thus every tube or line accesses the tank from the top. The bottom plate is slanted to minimize heel which is the residue of the previous transfer. This tank is designed not to yield any unmeasured volume and/or dead volume for circulation by employing an air-lift system instead of a pump.

Nominal volume of the tank is designed as 300 liter at 1200 mm liquid level. Compressed air is supplied to the dip-tube through a humidifier (a small cylindrical pot supplied by water). The air flows slowly downward at a rate around 6-7 liter/hour, and bubbles are formed and released periodically around every one second at the tip of dip-tube. Atmospheric pressure of the space above the solution is controlled a little (~ 0.5 kPa) negative to the atmospheric pressure of the room where ELTM and other tank-related instruments are installed. The tank is located in a cell on the basement floor, and ELTM is on the second floor, so the difference of altitude reaches ~ 11 m. Schematic diagram is shown in Figure 1.

It is important that dip-tube manometry is based on hydrostatic pressure measurement. However, plutonium nitrate solution in a tank is continuously stirred slowly to diffuse hydrogen gas resultant from alpha radiation. The solution flow around the tip of dip-tube affects the bubble formation and release, so it results in a large measurement bias as shown in Figure 4. It is necessary to stop stirring for a few minute or more to perform accurate measurement.

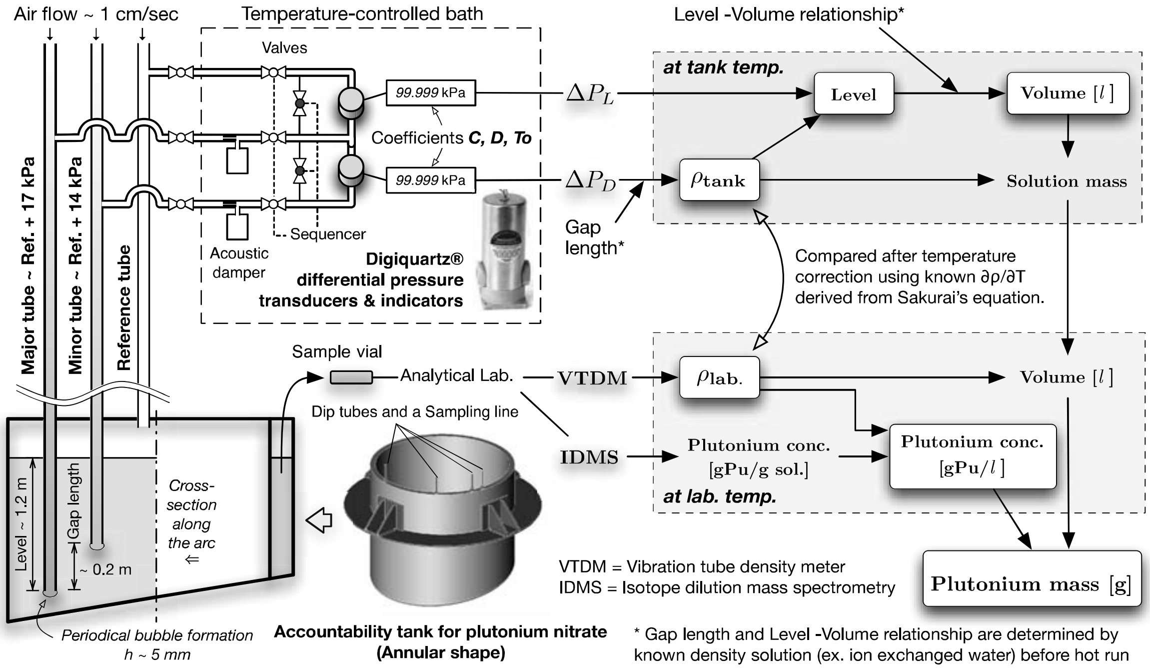


Figure 1 Schematic diagram of volume measurement system

2.2 Measurement of solution density and level

After the transfer, solution is homogenized by air agitation for an hour or more. Temperature of solution increases gradually after the transfer and almost saturates on the following day, then solution density, level and temperature are measured. The solution temperature was 34 °C to 40 °C which depends widely on isotopic composition. Slow stirring is temporary stopped during the measurement. Measured ΔP_L is proportional to the product of solution density and level. Measured ΔP_D is proportional to the product of solution density and vertical gap length between the tip of major tube and the tip of minor tube. The gap length is a tank specific constant, which is designed as 200 mm and determined at initial tank calibration using the reference liquid with known density. The initial tank calibration was carried out in advance of all volume measurements. Therefore, solution density is calculated from ΔP_D and gap length, then solution level is calculated from ΔP_L and solution density, that is the meaning to read the scale.

Typical ΔP_L and ΔP_D are 17 kPa and 3 kPa respectively for plutonium nitrate solution at nominal level, supposing the density of plutonium nitrate solution as $1.45\ \mathrm{g \cdot cm^{-3}}$. For water as the reference liquid, ΔP_L and ΔP_D are 12 kPa and 2 kPa respectively. Although ΔP_D is only one sixth of ΔP_L , no other transducer with a large full scale of 6 psi (41 kPa) could be applied. Thus relative measurement uncertainty of ΔP_D is six times larger than the one of ΔP_L . Roughly evaluated propagation of measurement uncertainty is shown in Figure 2 where relative weight for ΔP_L measurement uncertainty for plutonium nitrate solution is set to be one. The relative weights for the density and the weight of reference liquid are also supposed to be one because they should be the smallest ones. It is very important in Figure 2 that the relative uncertainty of ΔP_D at initial tank calibration and gap length as a consequence becomes the bottleneck of the propagation. Its uncertainty occupies three forth of the total one. To reduce volume measurement uncertainty to 0.1 - 0.2 %, it is necessary to reduce pressure measurement uncertainty to 0.1 % of the 2 kPa, that is 2 Pa.

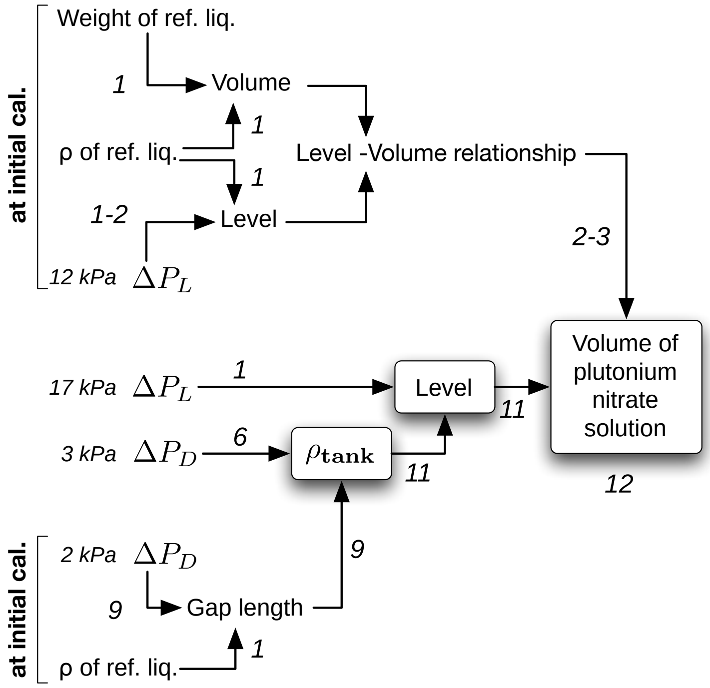


Figure 2 Roughly evaluated propagation of measurement uncertainty (relative)

2.3 Tank calibration and conversion of level to volume

Obtained solution level is converted to solution volume by a set of polynomial equations expressing level-volume relationship which is also tank specific. The relationship is determined at initial tank calibration using the reference liquid with known density and weight, in parallel with determining gap length. The level is calculated from ΔP_L and the density, and the volume is calculated from the weight and the density, that is the meaning to graduate the scale.

The initial tank calibration at PCDF was carried out in the presence of inspectorate in 1982, using ion exchanged air saturated water as the reference liquid. A prototype ELTM which was almost same as the current one except for automated functions and full scale of ΔP_L transducer was used. The initial tank calibration was carried out very carefully and prudently because it is very difficult to carry out tank calibration again with pure water after the start of hot operation. It is possible to carry out tank re-calibration using nitric acid to avoid polymerization of Pu(IV) at very low acidity, however it is difficult to obtain the equivalent measurement uncertainty to the one of initial calibration, because complete clean out is difficult to do and to be verified by inspectorate. Therefore, the details of initial calibration we have done are explained here. Key points of the procedure had been preliminary discussed and agreed with inspectorate.

First of all, residue of solution at test operation was removed. No solution was visually confirmed by operator and inspectorate from a hole at the top of tank, which was possible because the calibration was done at the final stage of facility construction. Ion exchanged air saturated water (~ 350 L) was prepared and put into a few pots on the previous day to stabilize temperature (~ 22 °C). Valves on the pipelines connected to internal pipes were set to the same condition as normal volume measurement condition. The purpose was to obtain reproducible condition of liquid volume inside the internal pipes. Air flow rate of dip-tube were also set to the same condition as normal condition. On the other hand, air flow for slow stirring was stopped to avoid fluctuation of measured pressure. An electro-balance specially reinforced to measure directly the weight of pot (~ 100 kg) was prepared and adjusted to 0.00 ± 0.01 kg using standard weights. ELTM was adjusted using a pneumatic dead-weight tester as a standard pressure. Average and standard deviation from the standard was 2 ± 2 Pa and 1 ± 1 Pa for ΔP_L and ΔP_D , respectively. The average was corrected at the time of data treatment to prevent an eternal bias on level-volume relationship or gap length.

Then, a line to feed the reference liquid to the tank was wetted to avoid any loss of the liquid at the first feed. After the wet, the pot was connected to the feed line, then a small volume of the reference liquid was fed into the tank using a small pump immersed in the liquid, followed by disconnecting the pot from the feed line. Weight measurements were performed before and after the feed. A power cable to the pump and a connecting tube were completely disconnected in every weight measurement, because any flexible suspension from an external support instead of disconnection causes a slight change in the force on the balance from the support which corresponds to the change in buoyancy acting on the cable and the tube from the liquid dependent on liquid level. Another buoyancy acting on the reference liquid and the standard weights from the surrounding air caused by differences of densities was corrected by an general equation.

After some waiting time, ΔP_L , ΔP_D and temperature were measured. The waiting time was set to avoid a small difference between the fed volume and actually increased volume at the bottom of the tank caused by slowly rolling water drops on the bottom plate just after the feed.

This work was repeated step-by-step until the tank was filled over the nominal level. Cumulative volume of the liquid in the tank was calculated from the sum of fed weight and the liquid density in the tank. The density was calculated from the chronological scientific tables and measured temperature. The density was also used to determine liquid level and gap length from ΔP_L and ΔP_D . At present, ITS-90 formula for air-saturated pure water propose by NIST is available(13). Measured temperature was 21.5 ± 0.5 °C throughout the calibration.

As a result, a discrete level-volume relationship and gap length were obtained at the room temperature. The same procedure was repeated three times to obtain dense measurement points of level and to decrease random measurement uncertainty. Finally, a set of polynomial equations (third order in maximum) expressing level-volume relationship was determined from precise fitting. Fitting error was less than 0.04 L at nominal level and less than 0.15 L throughout the level. Such precise fitting was achieved by an appropriate categorization of liquid level corresponding to the change in horizontal cross-section area evaluated as the first derivation of discrete dataset of volume-level relationship. Obtained boundary of the categorization was compared to the drawings of the tank. Statistical technique unfounded on the drawings was prudently not employed.

Obtained set of equations at the room temperature can be converted to the one at different temperature for volume measurement of plutonium nitrate solution, assuming isotropic tank thermal expansion and cubic expansion coefficient of tank material. However, such conversion was not employed in 1982, because the effect of tank thermal expansion was relatively small compared to the solution thermal expansion, moreover the assumption of isotropic thermal expansion had not been proved to be appropriate. Practical data treatment at present for tank thermal expansion and other measurement biases is explained in the section 3.3 & 3.4.

2.4 Sampling and analysis

To return to the procedure to determine plutonium weight, a sample is taken after the volume measurement from a sampling line where solution is circulated between tank and sampling equipment by air-lift system supported by vacuum. The tip of the sampling line lies at around half of the nominal level. Circulation is done half an hour or more in parallel with agitation, then several tens of cubic centimeter solution are taken and sent to analytical laboratory. Treatments/ preparations of the sample are carried out within a couple of days to avoid condensation. For plutonium concentration, isotope dilution mass spectrometry

(IDMS) is employed. For solution density, vibrating tube density meter (VTDM) is employed. Measurement uncertainty are 0.1 - 0.2 % and 0.05 % for IDMS and VTDM based on ITV, respectively. Results are reported with a volumetric expression typically [gPu•l^{-1}] and [g•cm^{-3}] where an appropriate base temperature, for example 31 °C, 35 °C or 37 °C were set by the specialist of analysis.

2.5 Temperature correction and plutonium weight determination

Total plutonium weight in the tank is given as a product of analytical result (volume-based plutonium concentration) and solution volume at the base temperature. The solution volume at the base temperature is calculated as a product of measured solution volume by ELTM and a factor for temperature correction. The factor is given as a fraction calculated as the density measured from ΔP_D at the tank temperature divided by the density analyzed at the base temperature ($\rho_{tank}/\rho_{lab.}$). This correction factor is the inverse of actual solution thermal expansion fraction.

This factor is indispensable in contrast with the factor for tank thermal expansion because the expansion coefficient of plutonium nitrate solution reaches to around 0.07% per degree in the wide range of solution properties. Figure 3 shows theoretical plot of $\partial\rho/\partial T$ (% of ρ) within the range of properties in our facility, evaluated from the first derivation of an empirical density formula of plutonium nitrate solution proposed by Sakurai (1991)(14) as a function of plutonium concentration, acidity and temperature. Temperature discrepancy between the volume measurement and the base temperature reaches several degrees or more, thus the factor reaches to 0.1 % or more.

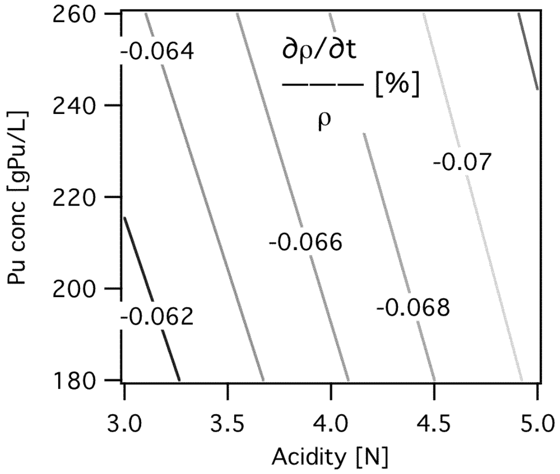


Figure 3 $\partial\rho/\partial T$ (% of ρ) of solution as the inverse of thermal expansion coefficient

This bias has been successfully corrected by the correction factor mentioned above, because an excellent consistency has been observed between the density by ELTM, the density by VTDM, the temperature discrepancy and the theoretical expansion coefficient, proved in the section 6.3. Correction by the temperature discrepancy and the theoretical expansion coefficient has not been adopted, because it includes another temperature measurement bias and/or uncertainty.

Meanwhile, a gravimetric expression (gPu for a unit weight of solution) instead of a volumetric expression is worth pausing to consider, because plutonium weight is simply given as a product of measured volume, measured density and plutonium concentration by gravimetric expression, where the product of measured volume and measured density is the solution mass which is independent from temperature. Gravimetric method has already been introduced to analytical procedure, thus the result obtained by the temperature correction is practically equivalent to the gravimetric method, that is the reason why actual solution thermal expansion fraction is used instead of theoretical solution thermal expansion fraction (0.07 % × temperature discrepancy).

2.6 Subtraction of heel to determine transferred plutonium weight

Finally, the weight of transferred plutonium is given as total plutonium weight subtracted by estimated plutonium weight in a small volume at the bottom of tank called heel which is the residue of the transfer of previous receipt solution to the storage tank. The weight of plutonium in the heel is a few hundreds of grams which corresponds several times larger than the target volume measurement uncertainty, thus it is necessary to measure or estimate precisely the volume and the plutonium concentration just after the yield of residue. They vary with time but their product is kept constant. The heel volume can be precisely measured from ΔP_L just after the transfer and ΔP_D just before the transfer. The plutonium concentration can be precisely estimated from the volume at sampling and the volume just before the transfer.

2.7 Comparison test of measured density and analyzed density

There are two solution densities at different temperature in the procedure shown in Figure 1. They are obtained from quite different sources : one is from pressure measurement and the other is from sampling and analysis. They should be equal at the same temperature if solution is well homogenized and sample is properly taken and treated. Thus, comparison of these densities is the proof of comprehensive accuracy and normality of measurements.

To compare two densities, density measured from ΔP_D at the tank temperature is adjusted to the density at the base temperature using a theoretical solution thermal expansion fraction. This fraction is calculated from the coefficient shown in Figure 3 and the temperature discrepancy. Normally, a constant 0.07 % per degree is used. If two densities do not match within the range of measurement uncertainty, it means something wrong or unexpected phenomenon happens. In such a case, investigation of the cause(s) and then re-measurement and verification have to be done immediately, because the solution should be stayed untouched until the completion of inspection though process operation continues. We have experienced such an unmatched case once, however the results of the test have been satisfactory which will be mentioned in the section 6.3.

2.8 Supplements

Formulae of solution density : Another formulae of solution density for plutonium nitrate solution as a function of plutonium concentration, acidity and temperature have been proposed by Maimoni (1979)(15), Kumar (1997)(16) and Charrin (2000)(17). The purpose of the formulae are, for example, to ensure the reliability of criticality safety design. Inter-comparison between three densities (ELTM, VTDM and the formulae) at the same temperature is very interesting to test normality and accuracy of measurement system and analytical results conclusively and comprehensively.

Liquid film remained on internal wall as an additional heel : We had been interested in a small portion of solution possibly remained on the surface of the internal wall (not the bottom) just after the transfer, because it might possibly cause unmeasured heel. However, it was concluded experimentally that such effect was negligible.

Capability of continuous solution monitoring : Measurement of solution mass by ELTM system as a product of measured volume and measured density is effective not only to apply the gravimetric method directly to plutonium mass determination but also to decrease fluctuation observed in a time-series data obtained by continuous solution monitoring(18). Figure 4 is an example of continuous monitoring where solution is slow stirred (normal condition), stirring is stopped and measured twice, then solution is circulated and sample is taken at IIV (Interim Inventory Verification), then returned to slow stirring.

Effect of evaporation appears commonly as a slight negative trend of volume and mass. Effect of stirring appears as a slight negative bias against volume (a.) and positive bias against density (a. and b.) all the time except for the short time when stirring is stopped. However, solution mass is almost free from the effect of stirring, and it is also free from the change in temperature before and after the sampling, although the volume changes around 0.1 liter according to the change in temperature around 0.6 °C. Sample volume is small compared to the effect of temperature on solution volume.

It is estimated that the bias during stirring is caused mainly by the solution flow around the minor tube which will be explained in Figure 8. Solution flow around the major tube and its effect on pressure measurement is estimated relatively to be small because the tip of major tube lies at the bottom of tank where solution flow is stagnated. Pressure of the major tube is proportional to the product of level and density which is close to mass, that is the reason why mass monitoring is almost free from the effect of stirring.

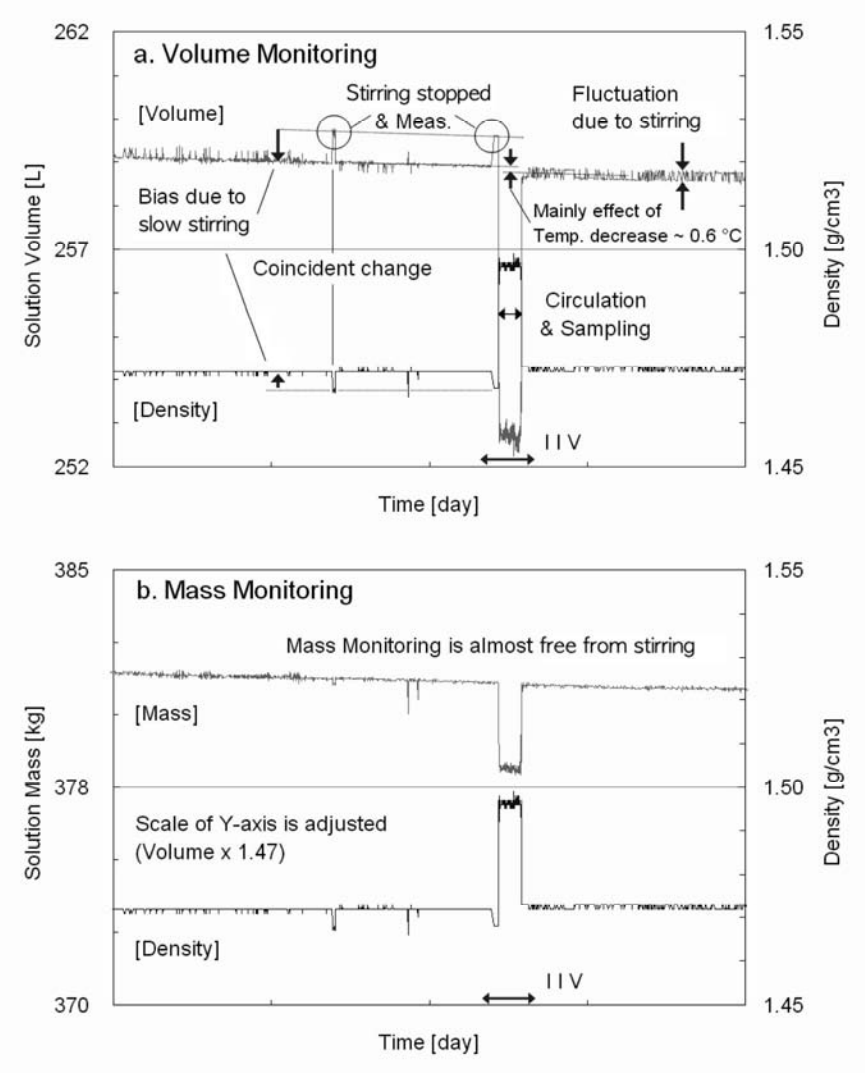


Figure 4 An example of continuous monitoring by volume and by solution mass

3. Measurement biases to be considered

Guidelines to achieve accurate initial tank calibration and volume measurement have been published as a series of ISO international standards from ISO 18213-1 to 18213-6 in the past few years(19). The original had been published as ANSI report N15.19 in 1989. Essential techniques in Japan had been developed at TRP and PCDF since the late 1970s to the first 1980s. ISO standard is very general, but a little apart from practical measurement procedure reflecting facility specific conditions. Initial tank calibration and volume measurement carried out at PCDF in 1982 was consistent with the standards though some corrections were evaluated by a slight different way or evaluated as negligible reflecting the structural and operational conditions of the facility.

There are five major sources of measurement biases to be considered to reduce volume measurement uncertainty to 0.1 - 0.2 % or pressure measurement uncertainty to less than 2 Pa : a) thermal expansion of solution ; b) air column effect ; c) thermal expansion of tank ; d) bubble effect ; and e) pressure loss. They are arranged in the order of magnitude in our facility. Another biases to be corrected by general methods are : a) buoyancy effect acts on weight measurement of the reference solution and b) local variation of gravitational acceleration (around ± 0.2 %) from the standard gravitational acceleration ($9.80665\ \mathrm{m \cdot s^{-2}}$) due to longitude, latitude, bouguer anomaly, elevation and altitude. Actual measurement or careful estimation of the gravitational acceleration is necessary.

Table 1 Measurement biases at typical conditions in PCDF

Source	Thermal expansion of solution	Air column effect	Thermal expansion of tank	Bubble effect	Pressure loss
Magnitude and condition	~ 0.42 % If diff. of temp. between vol. meas. and analysis is 6 °C	~ 0.13 % If diff. of altitude between tank and transducer is 11 m	~ 0.08 % If diff. of sol. temp. between tank cal. and vol. meas. is 16 °C	~ 0.05 % If inner dia. of dip-tube is 12.7 mm and thickness is 2.3 mm	~ 0.001 % If inner dia. of dip-tube is 12.7 mm and differential length is 3 m
Direction	Case by case	meas. < actual	meas. < actual	meas. > actual	meas. < actual
Correction	Done	Done	Not necessary in the condition above, but necessary if sol. temp. is higher		Not necessary

Sources, magnitudes, directions of measurement biases at typical condition in our facility are summarized in Table 1. For thermal expansion of solution and air column effect, corrections had been introduced since the start of operation. For bubble effect and tank thermal expansion, biases were precisely evaluated in the middle of 1990s and proved to be small but not negligible. Fortunately, it was also proved that over half of the effect of tank thermal expansion was in balance with bubble effect within current operational conditions. Corrections for these biases will be introduced at the time of high-burn up plutonium reprocessing in a next few years. For pressure loss, correction is not necessary in our facility.

3.1 Thermal expansion of solution

This source results in a bias several times larger than the target uncertainty, corresponds to the discrepancy between the temperature at volume measurement and the base temperature. This bias has been corrected by the temperature correction method as mentioned in the section 2.5. It is expected that the actual temperature correction factor calculated from two densities is equal to the inverse of theoretical volume expansion fraction calculated from temperature discrepancy and the coefficient. Results of the comparison test which will be mentioned in the section 6.3 is the proof of this expectation. Estimated error remained after the correction is 0.03 % as the mean of the test.

3.2 Air column effect

This source results in a bias a few times larger than the target uncertainty, corresponds mainly to the difference of altitude from tank to ELTM. Measured pressure is a little lower than the pressure at the tip of dip-tube, that is same as low atmospheric pressure at high mountain. A drawing and notations to derive equations to correct air column effect is shown in Figure 5. The air flows very slowly around 1.4 cm•sec^{-1}, so static approximation is available. Vertical distribution of air pressure $P(h)$ and air density $\rho(h)$ in each tube including the space above the solution is simply expressed by exponential curve :

$$\frac{dP(h)}{dh} = -\rho(h)g\ ,\ \rho(h) = \frac{W}{RT}\,P(h) \quad \Longrightarrow \quad P(h) = C\ \exp\left(-\frac{Wg}{RT}\,h\right)$$

where W is apparent molecular weight of air, h is altitude from the tip of tube A, and C is an integration constant. Differential pressure ΔP is given as :

$$\begin{aligned} P_A(h) - P_B(h) &= C_A\ \exp\left(-\frac{Wg}{RT}\,h\right) - C_B\ \exp\left(-\frac{Wg}{RT}\,h\right) \\ &= (\rho_{sol} - \rho_{air})\ g\ H_{sol} \times \exp\left(-\frac{Wg}{RT}\,h\right) \end{aligned}$$

where C_A is equal to the pressure at $h = 0$ of tube A, C_B is equal to the pressure at $h = 0$ of tube B which is equivalent to $h = 0$ of virtual empty vessel connected to the tank shown in Figure 5. The important premise is that the same temperature T and the same apparent molecular weight W are applied throughout the air in the tubes. This premise is satisfied because dip-tubes are passing through operator's work areas where temperature is well controlled, there is no high temperature apparatus near the tube, and water level of humidifier (or moisture content) are controlled. Furthermore, air density ρ_{air} is approximated to a constant 1.2 kg•m^{-3} under the fully humidified and pressurized condition, because variation of air density is ± 0.15 kg•m^{-3} that is negligible compared to solution density.

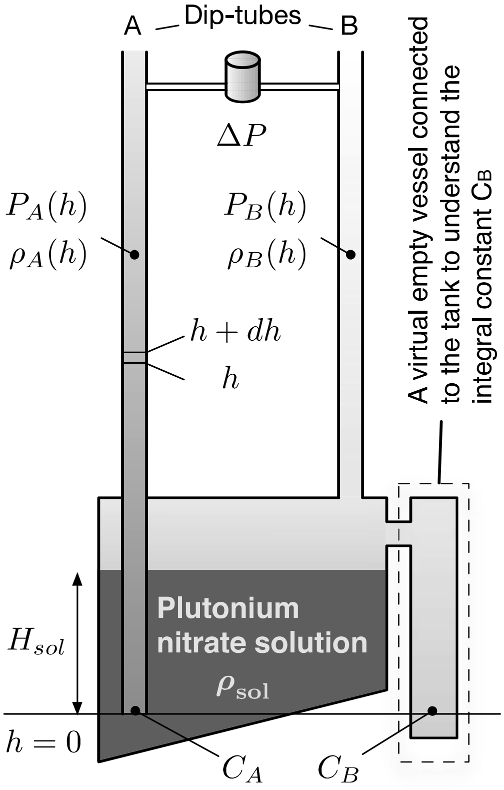


Figure 5 Schematic drawing and notations to formulate air column effect

Therefore, ΔP_L is simply given using SI units as :

$$\Delta P_L \quad \simeq \quad 0.99874 \times (\rho_{sol} - 1.2)\, g\, H_{sol}$$

where 0.99874 is a factor to correct air column effect which is calculated supposing W = 0.029 kg•mol^{-1}, R = 8.3145 J•mol^{-1}•K^{-1}, g = 9.80665 m•sec^{-2}, T = 298 K and h = 11 m. In the same way, ΔP_D is simply given using SI units as :

$$\Delta P_D \quad \simeq \quad 0.99874 \times (\rho_{sol} - 1.2)\, g\, L_g$$

Another approach and equations were derived and employed in ISO standard which could be applied to general situation. However the equations has too many variables to be practically applied. If T and W are simply supposed as we did, the ISO approach can be successfully reduced to the equations above. Despite a little modification of the factor, we have been employing this simple assumption and equations since the initial tank calibration. Estimated error remained after the correction is less than 0.03 % mainly as a result of assuming air density as a constant.

3.3 Thermal expansion of tank

This source results in a bias around half of the target uncertainty, corresponds to the expansion coefficient of tank material and temperature discrepancy between initial tank calibration and volume measurement. Temperature of plutonium nitrate solution at PCDF was 34 °C to 40 °C, and most frequent temperature was 36 °C to 38 °C. However, the level-volume relationship at the initial calibration had been determined at 21.5 °C. Cubic expansion coefficient of tank material is 0.005 % per degree (equivalent to linear expansion coefficient of austenitic stainless steel as 0.0017 % per degree), so the effect of tank thermal expansion reaches to 0.08 % for 16 degrees. The important premise is that isotropic thermal expansion is assumed despite its welded construction and rigid supports to concrete cell.

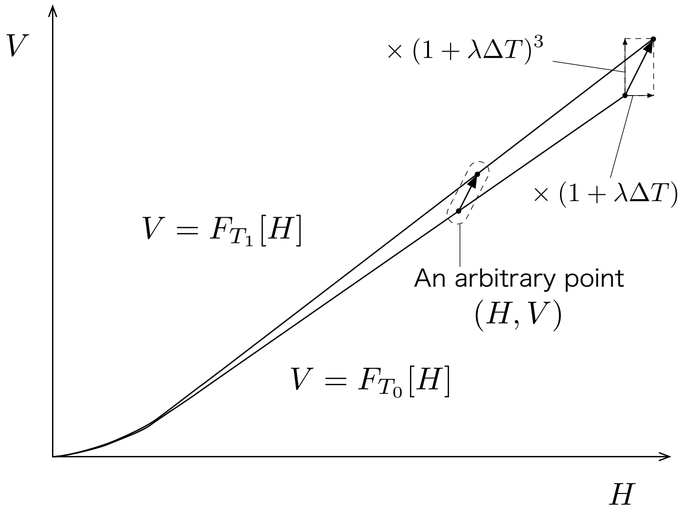


Figure 6 Notations to formulate effect of tank thermal expansion

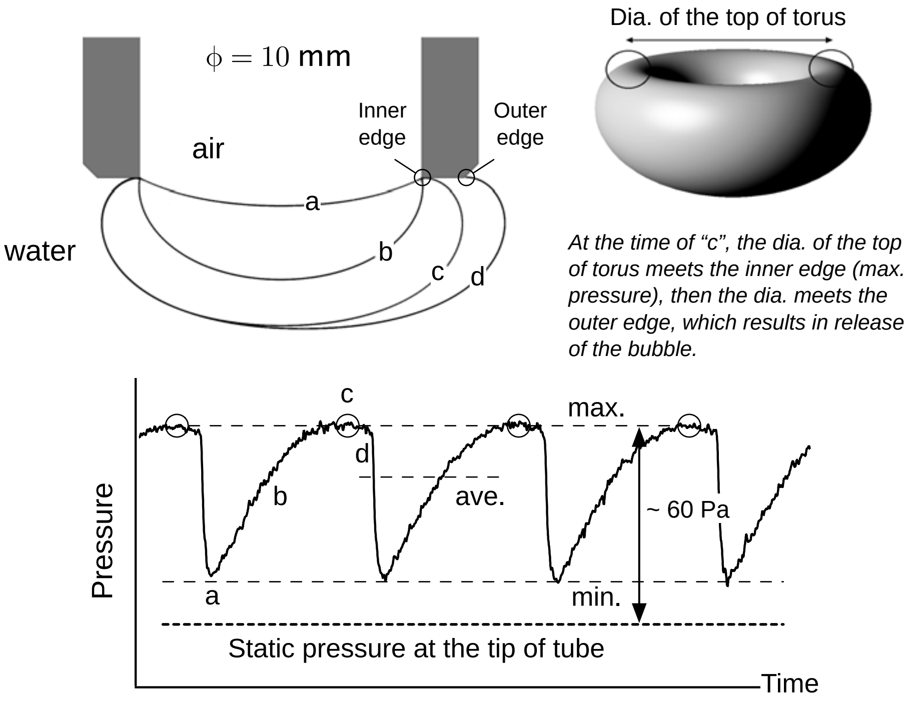
In contrast with the biases due to solution expansion or air column effect, it is not so easy to understand whether this bias is positive or negative. It is helpful to imagine that the ideal solution with zero thermal expansion is filled in a cylindrical tank, and level-volume relationship which is a graduated scale expands thermally. If the tank expands, actual level decreases due to expansion of horizontal cross-section area by a factor of surface expansion, then the level is read by the expanded scale by a factor of linear expansion. Therefore, the obtained volume is smaller than the calibrated volume by a factor of cubic expansion. A drawing and notations to understand the change in level-volume relationship is shown in Figure 6. General expression of the change in level-volume relationship is given as :

$$V = F_{T_1}\left[H\right] \quad \rightleftharpoons \quad \frac{V}{(1+\lambda\Delta T)^3} = F_{T_0}\left[\frac{H}{(1+\lambda\Delta T)}\right]$$

where $\Delta T = T_1 - T_0$ and λ is linear expansion coefficient of tank material. Based on the equations above, level-volume relationship can be converted to another temperature. Availability of this assumption was obtained in Japan by a LASTAC project(20) to test and confirm accountability of a tank used for large scale reprocessing, which was carried out by Nuclear Materials Management Center. This simple assumption is also employed in ISO.

3.4 Bubble effect

This source results in a bias around half of the target uncertainty, corresponds to the sum of pressure increase caused by vertical shift of liquid-gas interface from the tip of tube and additional pressure caused by curvature of the interface at the bottom. The static axisymmetric shape of the interface is calculated numerically from the force balance equations and a special coordinate system(21).



At the time of "c", the dia. of the top of torus meets the inner edge (max. pressure), then the dia. meets the outer edge, which results in release of the bubble.

Figure 7 An example of experimentally observed pressure wave in water

For example, in water with surface tension of 0.072 $N\cdot m^{-1}$ at room temperature, the bubble height around 5 mm yields around 50 Pa and the curvature around 100 m^{-1} yields around 14 Pa according to the Young-Laplace equation. An example of pressure wave observed in water near the tip of tube by high speed pressure transducer is shown in Figure 7. Pressure varies within the range from 10 to 60 Pa higher than the hydrostatic pressure at the tip of tube, which is called overpressure.

The overpressure is recognized to be composed of a constant bias from hydrostatic pressure (the integrated average "ave.") and a fluctuation around the bias. The fluctuation can sufficiently be reduced by a pneumatic damper mentioned in the section 4.4, so it is necessary to evaluate the constant bias. Actual pressure wave observed at the end of dip-tube in the room where ELTM is installed is quite different due to acoustic effect of the tube, however the integrated average is kept not changed at the end. It is important that the bias does not affect directly the volume measurement but the difference of the biases between in water at tank calibration and in plutonium nitrate solution affects volume measurement.

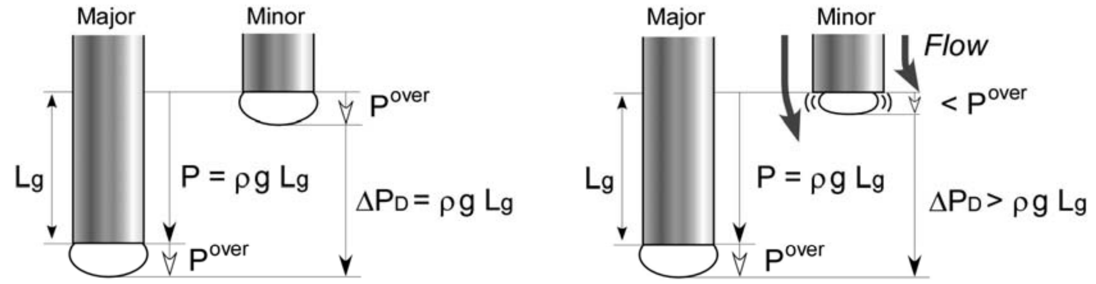
Static axisymmetric shape of the liquid-gas interface had been formulated and numerically calculated in the late nineteenth century by Bashforth and Adams(22), then studied methodologically by Hartland and Hartley(23). However the specific shape of the torus can only be obtained by the special coordinate system. The force balance equations contain physical properties in the form of $\Delta\rho g/\sigma$ where σ is surface tension and $\Delta\rho$ is differential density between liquid and gas, and the equations are independent from solution depth. Experimentally observed stable shape in water was independent from solution depth indeed, and the shape was very close to the calculated one, because bubble grows slowly enough to satisfy the condition that the force of surface tension is larger than the force of inertia. Based on the observation, it was found that the max. pressure and the bubble release were dominated by the relation between the torus diameter and the inner/outer diameter of the tube. It was also found that the bubble shape in the solution with the same $\Delta\rho g/\sigma$ as plutonium nitrate solution (such as ethanol - water mixed solution) became more flat than in water, which was close to the numerical expectation. As a result, we have developed a new method to evaluate and correct the constant bias.

It is important that measurement of ΔP_D is independent from the bubble effect because the effects are almost same at major tube and at minor tube owing to very small difference of $\Delta\rho$ (0.03 $kg\cdot m^{-3}$) compared to $\Delta\rho$ (~ 1450 $kg\cdot m^{-3}$). In contrast, measurement of ΔP_L is affected by the bubble.

There are two ways to correct this bias in ISO standard. One is slow bubbling technique and the other is fast bubbling technique. The former can be utilized for analytical measurements (state of art) and the latter is for industrial operations (state of practice). In the case of slow bubbling where bubbling frequency is every half minute or less, maximum of the pressure variation ("c" in Figure 7) is measured and the overpressure at this point is evaluated and corrected. This evaluation is based on the book by Hartland and Hartley. In the case of fast bubbling where bubbling frequency is normal or around every one second, the integrated average is measured then the average of overpressure is evaluated and corrected. However, ISO standard for the fast bubbling is not based on the classic knowledge since the last century and a little apart from experimental results. Our conclusion was 8 Pa as the difference of constant biases on ΔP_L measurement in our facility(24).

This bias means additional 8 Pa is required to release a bubble in plutonium nitrate solution compared to in water. Therefore, measured volume of plutonium nitrate solution is larger than the calibrated volume around 0.05 % (8 Pa of 17 kPa). This bias is opposite to the bias by tank thermal expansion evaluated as 0.08 %. Therefore, over half of the effect of tank thermal expansion is in balance with the bubble effect within current operational conditions. Estimated error remained after cancellation is around 0.03 %.

It is very important that these results are based on the supposition that there is no solution flow near the tip of tube. We had observed a bias around + 5 Pa in ΔP_D measurement during slow stirring. It was estimated that overpressure necessary to release a bubble at the tip of minor tube becomes a little smaller owing to the perturbation caused by solution flow, though overpressure at the tip of major tube does not change because the solution is almost stagnated at the bottom, shown in Figure 8.



a) No solution flow b) Flow around minor tube

Figure 8 Effect of solution flow around minor tube on ΔP_D

3.5 Pressure loss

This source results in a bias much smaller than the target uncertainty, corresponds mainly to inner diameter and differential length of dip-tubes. As mentioned in the section 2.1, the air flow rate is 6-7 liter/hour that is equivalent to around 14 mm/sec. Therefore, the pressure loss in each tube can be calculated from the Hagen-Poiseuille law applied to laminar airflow in a tube with smooth inner surface :

$$Q = \frac{\pi a^4 \Delta P}{8 \mu l}$$

where Q is airflow rate (6-7 liter/hour), a is inner radius (6.35 mm), μ is viscosity (18 μPa･s), l is tube length and ΔP is pressure loss. Viscosity of air is almost independent from pressure. It is essential that pressure loss is proportional to the tube length and inversely proportional to the fourth power of inner radius. For this airflow at room temperature, Reynolds number is evaluated to be 11-14, thus laminar airflow is properly supposed.

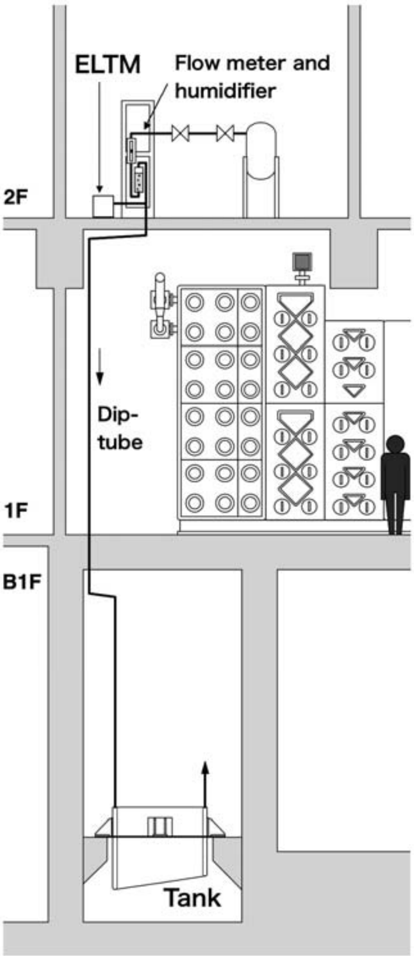


Figure 9 Layout of ELTM, dip-tubes and a tank

Differential tube length inside the tank is 1.4 m and 0.2 m for ΔP_L and ΔP_D, respectively. Differential tube length outside the tank (from the tank to ELTM) is estimated to be within 1.5 m, because two transducers are installed in the same room located almost above the tank and dip-tubes are installed in parallel as shown in Figure 9. Total differential tube length is estimated around 2.9 m and 1.7 m for ΔP_L and ΔP_D, respectively. As a result, pressure loss of ΔP_L and ΔP_D are estimated around 0.15 Pa and around 0.09 Pa, respectively. In addition, it is important that the pressure loss is almost independent from pressure itself in a tube because of the characteristics of viscosity. Therefore, pressure loss at volume measurement is almost in balance with the pressure loss at tank initial calibration. Variation of airflow rate can affect pressure loss which is ~ 10 %, so the effect of pressure loss is estimated less than 0.001 % in our facility.

However, the results would change if tube radius were 3 mm and differential tube length were 10 m, because pressure loss would reach to 10 Pa and its 10 % were 1 Pa that could not be negligible.

3.6 Uncertainty related to bias corrections

In conclusion, the uncertainty related to bias corrections is evaluated to be less than $0.052\% = \sqrt{0.03^2 + 0.03^2 + (0.08 - 0.05)^2 + 0.001^2}$ which will be used as type B uncertainty in the last section 6. This is not a constant bias but categorized to be the component of systematic character of measurement uncertainty.

3.7 Supplement

There is a supplement for thermal expansion of solution and temperature correction. If tank is exactly cylindrical which means that level-volume relationship is linear and passes through the origin, ΔP_L becomes exactly proportional to solution mass and insensitive to solution temperature. In such a case, another simple method is available where measured density from ΔP_D can be substituted with analyzed density at base temperature throughout measurement procedure. Actual tank shape is not cylindrical, however it is essential that solution mass as a product of volume and density measured by dip-tubes is theoretically not so sensitive to solution temperature. If solution density measured from ΔP_D has a little positive bias, it results in a little negative bias to solution level and volume, thus it results in very small bias to solution mass. That is the reason why we employ the actual temperature correction factor instead of the inverse of theoretical volume expansion fraction to which measurement uncertainty of solution temperature affects.

4. Electro-manometer system

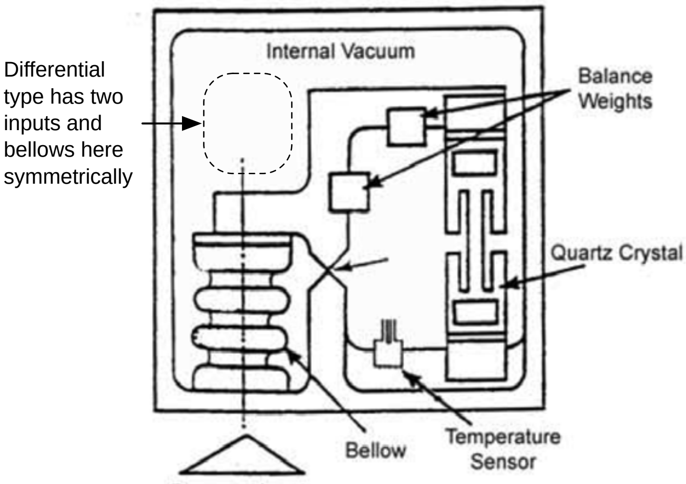
There are four ELTM systems for input tank, storage tank, plutonium-uranium mixing tank and mixed solution storage tank. They are independent but their shape is identical. The system is not a device temporary connected for material accounting but a device always connected for both material accounting and process monitoring purposes, because the system is useful to monitor very precisely the balance of volume or solution mass at solution transfer between tanks, at sampling and by natural evaporation, and to confirm normality of process operation.

Therefore, ELTM system was designed to be always ready to be used and to indicate solution density, level, volume, and status of the system on a real-time basis. This design was also effective for inspection frequently done or possibly done without notification in advance. Accessibility and verifiability by inspectorate were also introduced to the design, because the system had also been used by inspectorate, until installation of their own system, as an inspection device based on a premise that the device is carefully verified using their own standards and/or other approaches. For such an implementation, a precise procedure was negotiated and agreed in advance between operator and inspectorate to satisfy both inspection requirements and operation requirements.

The target measurement uncertainty for volume was set at 0.1 - 0.2 % and the one for pressure measurement was set at 2 Pa as one sigma, as described in the section 2.2. It was necessary that the targets should be satisfied continuously.

4.1 High precision differential pressure transducer

Quartz crystal was employed as a sensing element of the digiquartz® differential pressure transducer. The resonant frequency of oscillation varies with the stress induced by pressure. The mechanism to induce the stress is shown in Figure 10, where differential pressure acts on the effective area of the bellows to generate a force and torque about the pivot and compressively stress the resonator. The change in frequency of the quartz crystal oscillator is a measure of the applied pressure. Displacement of the quartz crystal is quite small which is in sharp contrast with conventional diaphragm manometers or bourdon tubes. As a result, remarkable repeatability, low hysteresis and excellent long-term stability as a feature of quartz crystal are realized. The output is detected with oscillator electronics similar to those used in precision clocks and counters. Variation of resonant frequency is not linear to pressure, so an equation and constants to convert frequency to pressure is provided by the manufacturer. The resonant frequency is 36 kHz and 40 kHz when full scale differential pressure is applied and no differential pressure is applied, respectively. Full scale of the currently used transducer at PCDF is 6 psi or 41 kPa for both ΔP_L and ΔP_D .



Differential type has two inputs and bellows here symmetrically

Quoted from the web of Paroscientific, Inc.

Figure 10 Mechanism of digiquartz® pressure transducer

Equation to convert frequency to pressure is given as :

$$P = C\left(1-\frac{T_0^2}{T^2}\right)\left[1-D\left(1-\frac{T_0^2}{T^2}\right)\right]$$

where P is pressure, T is period of oscillation, T_0 is a constant corresponds to the period of oscillation when P = 0, C and D are constants determined to minimize discrepancy between P and actual pressure applied as a reference. Frequency-pressure conversion by the equation above and primary data treatments are performed by microprocessor-based intelligent electronics. The transducer is not so adequate to observe high speed phenomenon because it takes a time to count frequency. The digiquartz® pressure transducer is the product of Paroscientific, Inc. in USA(25). The microprocessor-based intelligent electronics is the product of Tsukasa Sokken Co., Ltd. in Japan(26).

The announced accuracy, repeatability and resolution are 0.02 %, 0.005 % and 0.0001 % of full scale, respectively. The constants provided by the manufacturer are optimized for the full scale, however the actual measurement range is half or less of the full scale. In addition, the constants might vary dependent on aging characteristics of the vibrating quartz crystal. Therefore, a dead-weight tester to supply reference pressures was installed beside ELTM, then the constants were optimized by ourselves. Residual from the reference pressures could be optimized less than 0.5 ± 1 Pa as one sigma within the actual range. The residual is not a constant but a variable dependent on applied pressure. Furthermore, ± 1 Pa is combination of the repeatability of the transducer and the reference.

Besides, the resonant frequency of oscillation is also a function of temperature, so a temperature sensor to compensate the equation is embedded inside the transducer. However, we have selected to install the transducers inside a large temperature-controlled (± 1 °C) water bath instead of this compensation to stabilize physically the temperature of the transducer. Meanwhile, consideration for the effect of acceleration that is indispensable when the transducer is set on moving or flying object is not necessary.

4.2 Dead-weight tester as a reference

A pneumatic dead-weight tester which can generate discretely the reference pressures at 3, 5, 10, 15 and 20 kPa was employed. The tester consists of precisely fabricated piston-cylinder and weight disks. Mass of the discs and cross-section area of the piston-cylinder at a certain temperature were determined and certified by superior standards which were traceable to the national standards. The true reference pressures were calculated by ourselves from the certificates using local gravitational acceleration measured or well estimated, vertical level difference between the transducer and the tester, and temperature of piston and air.

Photo 1 Dead weight tester inside a rigid closed clean box

The tester is very sensitive to fluctuation of atmospheric pressure, dew formation, tilt of the tester and micro dust invasion to the tester. They disturb easily the smooth rotation of piston-cylinder, although the smooth rotation at a certain stable floating level over one minute at the minimum reference pressure is the evidence of normal working condition. To keep smooth rotation, the tester was installed inside a rigid closed clean square box shown in Photo 1 set beside ELTM in the same room where humidity and temperature were controlled. This box can shut off flutter of atmospheric pressure which is peculiar to nuclear facility where atmospheric pressure is negatively controlled. The minimum reference pressure was set higher than the lowest generated pressure in order to ensure the rotating time sufficiently.

The tester has been used not only to determine C, D constants but also to calibrate transducer once or twice a month not to determine these constants but to confirm or to verify the accuracy of pressure transducer before important measurement. Repeatability of the tester itself is not known but combination with the repeatability of the transducer is ± 1 Pa, which is difficult to be reduced due to its mechanical structure and facility's negative pressure control. Dead-weight tester is the product of DH-Budenberg Ltd. in United Kingdom(27), with a certificate by National Measurement Accreditation Service (NAMAS) or United Kingdom Accreditation Service (UKAS).

4.3 Sequential valve control to adjust zero point and calibration

Variation of zero point by temperature is the most worried source of measurement error, because volume measurement is carried out continuously without taking the transducer out from the conducting pipes. Periodical adjustment of zero point is indispensable although temperature of the transducer is stabilized by the water bath. In addition, occasional confirmation of zero point is necessary before important measurement to show inspectorate clearly that no bias exists. Compensation by a temperature sensor embedded in the transducer is effective to downsize the measurement system but it is not enough to ensure and to assure zero point.

Therefore, an automated circuit consists of five electric valves and a sequencer was installed on the conducting pipes to isolate physically transducers from dip-tubes and to adjust zero point. Objectives to be solved were influence of heat from electric valves and influence of enclosed pressure after isolation on zero point. As a countermeasure against heat, magnetic latch valves driven by permanent magnets and pulse current were employed to reduce heat. Simple two way valves (alternative of open or close) were employed to ensure no leakage from the valve. As a countermeasure against enclosed pressure, a sequential control of valves to release the pressure was introduced. If transducers are isolated simply by closing three valves at the same time, the pressure in the pipes to major, minor and reference are + 17 kPa, + 4 kPa and a little negative to the atmospheric pressure of work area, respectively. If both inlets of the differential transducer is simply shunted, the enclosed pressure (around middle of the above) causes a shift of zero compared to the normal zero adjustment where the pressure is atmospheric, although the differential pressure is truly zero. This characteristics depends on each transducer and corresponds to the enclosed pressure. Therefore, a sequence shown in Figure 11 was invented to release the enclosed pressure safely to the reference dip-tube before zero adjustment. As a result, zero point became to be adjusted always under the constant condition where the enclosed pressure was a little negative but close to atmospheric, which resulted in practically no variation of zero point.

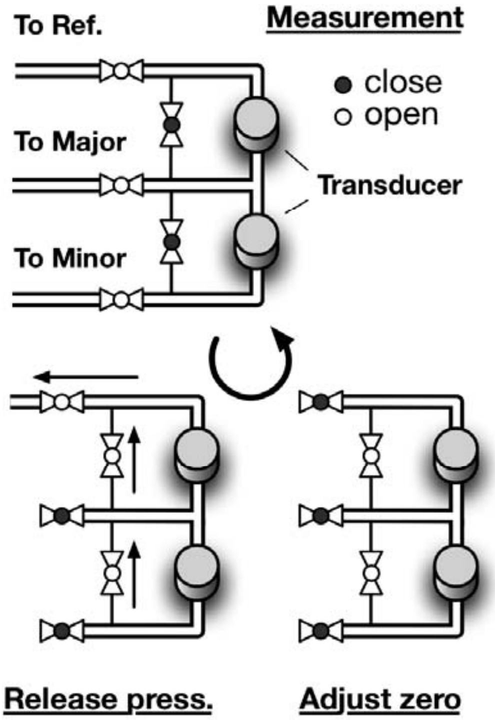


Figure 11 Sequential valve control for zero adjustment

This sequence has been used not only to adjust zero point but also to connect safely the dead-weight tester for periodical/occasional calibration without taking the transducer out from the conducting pipes, because the negative pressure is effective to prevent diffusion of contamination inside the conducting pipe that is rare but possible. A similar sequence including operation of manual valves in the boundary between ELTM and dip-tubes is utilized as a preparation of annual maintenance to ensure safety.

4.4 Pneumatic damping of pressure oscillation

Pressure at the tip of dip-tube is oscillating around every one second as shown in Figure 7. This pressure wave is transformed and attenuated at the end of dip-tube (at the conducting pipe) into a wave oscillating around the integrated average with a smaller range. The attenuation is a result of a time constant calculated from acoustic inertance and acoustic resistance which correspond to inductance and resistance of an electrical circuit. The time constant is around 0.4 sec at PCDF, and the range of actually observed oscillation is around half of the original.

However, the range was still larger than the target uncertainty by a factor of ten. Therefore, an acoustic low-pass filter consists of a needle and a small tank was attached on the conducting pipes to major and to minor. The needle acts as an acoustic resistance R_A and the tank acts as an acoustic compliance C_A which is equivalent to capacitance of an electrical circuit.

$$R_A = \frac{\Delta P}{Q} \simeq \frac{8\,\mu\,l}{\pi\,a^4}$$

$$C_A = \frac{V}{\rho\,c^2}$$

where μ is viscosity (18 μPa•s), ρ is density (1.2 kg•m^{-3}), c is speed of sound (347 m•s^{-1}), a is inner radius of the needle, l is length of the needle, V is volume of the tank (1 liter). Definition of R_A is approximated by Hagen-Poiseuille law, because the oscillating frequency is low. The time constant calculated as a product of R_A and C_A was adjusted to be 1-2 sec using an appropriate needle used for injection (e.g. 21 G needle has 0.51 mm inner diameter and $a \sim 0.25$ mm) and $l \sim 20$ mm. As a result, oscillation was sufficiently reduced to one tenth or less of the input, thus the oscillation was reduced to several pascals.

To obtain smaller oscillation, the output of transducer was moving averaged. The bubbling frequency is around once per second, so the averaging time was set normally at 5-15 seconds. As a result, fluctuation due to oscillation was sufficiently reduced to around 1 Pa.

4.5 Data acquisition and store

Measured pressure, the output of microprocessor-based intelligent electronics of the transducer, is always indicated by the same electronics on a real-time basis. Outputs from 8 transducers for 4 tanks are acquired by a PC every one minute for each tank, thus acquisition is switched every 15 seconds for each tank. Solution density, level and volume are calculated by the PC and displayed on the PC accompanied by raw pressure data. Calculated solution density is stored for re-use when solution level becomes lower than the tip of minor tube by operation and/or natural evaporation. Such situation is easily detected by comparing ΔP_L and ΔP_D because they are same in such a case. Calculation of solution level and volume continues in such a case using the stored density.

The PC also controls the valve sequencer and detects its anomaly. All acquired data, calculated values, anomaly information such as a fault of the sequencer or a deviation from normal range of pressure fluctuation are recorded every one minute. The PC is battery backed up and all information can be stored safely at power failure. Transducers and microprocessor-based intelligent electronics are connected to uninterruptible power source. Recorded data can be exported to another PC to be transformed to a time trend chart and/or to be analyzed. Data acquisition system and related software are designed, manufactured and totally assembled with sequential valve control system by Tsukasa Sokken Co., Ltd.

4.6 Expected measurement uncertainty in design base

The random component of pressure measurement uncertainty by ELTM system was evaluated to be the square-root of the sum of squares of the following uncertainties : 1 Pa due to pressure oscillation ; 1 Pa due to deviation of zero point and/or variation of temperature sensitivity ; and 1 Pa due to combination of the repeatability of the transducer and the reference. Therefore, the value 2 Pa was prudently expected as one sigma of the random component. To ensure this expectation, the same value 2 Pa was set at as a normal permissible limit of discrepancy from the reference at calibration by the dead-weight tester. For the component of systematic character, a small discrepancy less than 0.5 Pa between the reference and the transducer lay after the optimization, which was less than 0.017 % of 3 kPa (minimum reference) used not as a constant bias but as type B uncertainty in the last section 6. This systematic character was sufficiently small nearly one forth of the random component.

In conclusion, the random component of relative measurement uncertainty of ELTM system as one sigma was expected in design base to be 0.012 % of 17 kPa for ΔP_L measurement of plutonium nitrate solution, 0.067 % of 3 kPa for ΔP_D measurement of plutonium nitrate solution, 0.017 % of 12 kPa for ΔP_L measurement of reference liquid, and 0.100 % of 2 kPa for ΔP_D measurement of reference liquid. It should be noted that the last one becomes the bottleneck of uncertainty propagation. The total relative uncertainty of random component was expected according to Figure 2 to be 0.1 - 0.2 % as one sigma which was consistent to the target uncertainty of the development and nearly the same as analytical measurements used for plutonium accounting.

5. Operation and maintenance performed over fifteen years

The first transfer of plutonium nitrate solution from TRP to PCDF was carried out in 1983. In prior to the first transfer, initial tank calibration was carried out in 1982. Since then, the transfer has been carried out 124 times by the end of 2009. Cumulative throughput of plutonium has reached to 6.4 tons which has been determined by ELTM system and destructive analysis. The system has also been used for inventory determination or evaluation. Inventory determination accompanied by interruption of process operation and clean-out operation has been carried out twice (until 1989) or once (since 1989) a year. Inventory evaluation during operation has been done at monthly base. Moreover, re-calibration of input tank which was similar to the initial tank calibration but nitric acid was used instead of pure water has been carried out every year to re-confirm the consistency of gap length and level-volume relationship.

The purposes of the demonstration were to keep the expected volume measurement uncertainty in design base described in the previous section by appropriate maintenances and to prove it to be practical as industrial operations. To achieve these purposes, daily check, monthly/occasional calibration, and annual maintenance have been carried out to control pressure measurement discrepancy from the reference within the permissible limit normally set at 2 Pa described in the previous section. Besides, half-clogging at the tip of dip-tube was unavoidable and necessary to be treated, although the air put into dip-tube was well humidified. In addition, unexpected replacement of transducer was experienced and the system was improved to reduce the time for replacement and not to require preventive maintenance. A tool to diagnose the replacement was found.

5.1 Daily check

Air flow rate to the dip-tubes, water level of the humidifier, water level and temperature of the water bath, and indications of ELTM system were checked daily. The indications were compared with the expectations derived from the most recent indications and the operational records afterwards, considering loss of volume by natural evaporation normally around 0.1 liter or more per day at PCDF. This comparison was very useful to find unexpected situation such as occurrence of half-clogging in the dip-tube or malfunction of ELTM system. Speed of natural evaporation was different for each tank mainly due to ventilation flow, but almost independent from solution level due to the cylindrical shape of tank.

5.2 Treatment of half-clogging

Although compressed air put to the dip-tubes was well humidified, it was difficult, sooner or later, to avoid occurrence of half-clogging. Increase of ΔP_L and

ΔP_D due to half-clogging of major tube, decrease of ΔP_D due to half-clogging of minor tube, and their combination brought rapid and accelerating but not sudden change in density, level and volume. Examples are shown in Figure 12. Gradual shift of pressure and gradual growth of fluctuation are observed. The change usually takes several hours and clearly be distinguished as an anomaly. It is impossible to observe visually the clogging, however it is possible to estimate roughly the hole size according to the rule by Hagen-Poiseuille where pressure loss is in proportional to the inverse of fourth power of the hole diameter. It is also estimated that the clogging lies not at the tip but a little above from the tip where the liquid-gas interface fluctuates and precipitation occurs. In our experience, the effect of clogging on level-volume relationship and gap length seems to be small and random as described in the later section, which means the clogging lies not at the tip and not cumulative. However, an appropriate treatment is necessary immediately when anomaly is observed, despite the effect of clogging is small and difficult to be detected at early-stage.

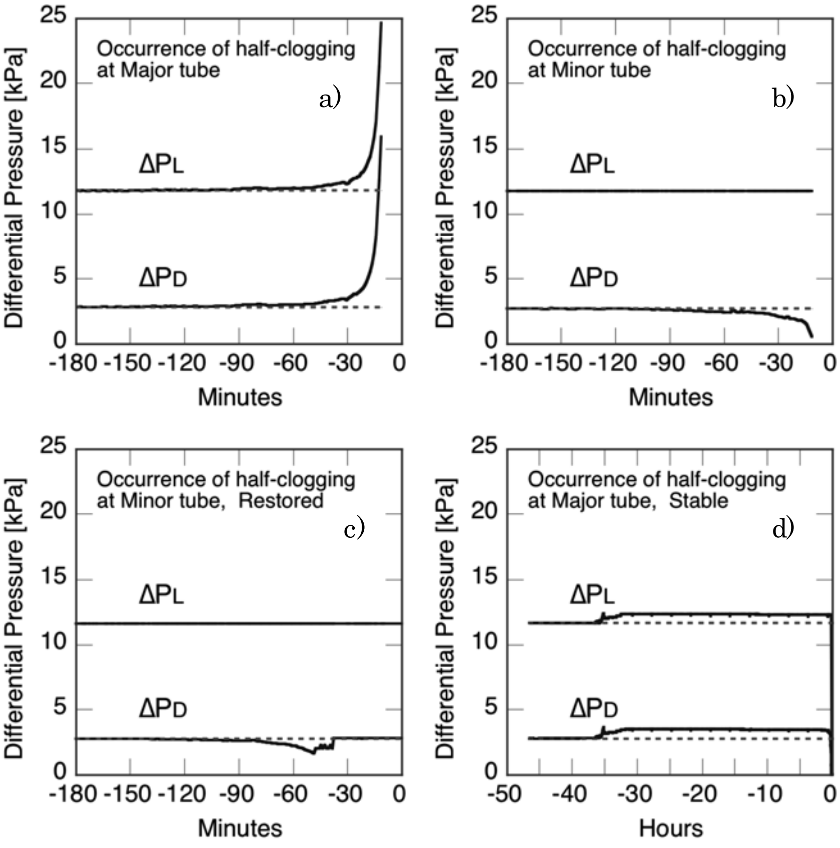


Figure 12 Examples of half-clogging

The half-clogging showing anomaly can be treated, in our experience, by putting nitric acid and compressed air sharply several times at a certain flow rate, keeping the reference pressure to be negative. It is also effective that the half-clogging is immersed to nitrate solution followed by putting compressed air sharply. However, it was difficult to avoid recurrence of half-clogging. More effective and efficient method is under development.

5.3 Monthly and occasional calibration

Comparison test with reference pressure to confirm accuracy has been carried out once a month normally at the end of month. This comparison test has also been carried out before important measurement and/or inspection such as receipt of plutonium, inventory determination or evaluation and other regular/occasional inspections. The test result lay normally inside the permissible limit and the constants C and D remained normally not changed. Obtained results at inspection were shared between operator and inspectorate.

In prior to connect dead-weight tester to ELTM system, the system has to be isolated from the dip-tubes. The procedure of isolation is very important to control contamination inside the conducting pipe which is rare but possible. In order to avoid scattering of the contamination to work area, internal pressure of the pipe was turned lower than the atmospheric pressure of work area. To turn the pressure, the air inside the pipe was released to the reference line by the same valve control sequence as zero-adjustment. Then, three valves on the conducting pipes and two valves for shunt were kept closed during calibration. It usually took about half an hour to complete calibration.

5.4 Annual maintenance

Annual maintenance has been carried out by a company specialized in precise treatment of dead-weight tester and digiquartz® transducer normally by the company which manufactured totally the ELTM system. Before the annual maintenance, information related to malfunctions or faults experienced since the last maintenance were provided to the company. If necessary, improvements were proposed by JAEA and/or the company and discussed. Based on the provided information and the results of discussion, the company proposed a preliminary plan of maintenance. The plan was checked and approved by JAEA, then informed to the government and IAEA in advance of the maintenance.

Before the start of maintenance on the first day, the system has to be isolated from dip-tubes by JAEA operator according to the same manner as monthly calibration. Instead of controlling three electric valves on the conducting pipes, three manual valves located on the boundary between ELTM system and dip-tubes are carefully controlled, and the air in damping tanks is safely released to the reference line. After proper preparations to avoid contamination, three closing plugs set beside the manual valve are carefully opened. Contamination inside the conducting pipe at the boundary is checked by a cotton swab. After the completion of isolation and contamination check by JAEA, the company starts maintenance. Fortunately, we have no experience of contamination not only at annual maintenance but also at monthly/occasional calibration.

Mechanical components such as electric valves are dismounted, taken apart and checked visually. Then, each parts are cleaned up and consumable parts such as gasket and plunger are exchanged. Finally, the parts are build up and behavior of the component is checked. Visual check is performed carefully not only for mechanical components but also for static components such as pipes and a water bath. Electric components such as valve sequencer, microprocessor-based electronics and PC are tested and/or exchanged if necessary. Batteries for uninterruptible power source are exchanged periodically according to manufacturer's suggestion.

To confirm no leakage from conducting pipes and valves after building up, compressed air of 120-140 kPa is gradually injected to the pipes. It is necessary to wait for ten minutes to release adiabatic compressive heat due to injection. After the ten minutes, variation of pressure is observed for five minutes. Permissible range of the variation is set at one percent, because it varies not only by leakage but also by temperature fluctuation of the enclosed air. Pressure measurement is not so sensitive to the small leakage between conducting pipes and working area compared to the leakage between pipes.

Dead-weight tester is also taken apart, checked visually, cleaned up and build up. Accuracy and normality of the tester after building up is assured by rotating the piston-cylinder smoothly over one minute at a certain stable floating level at the minimum reference pressure (3 kPa). Performance of the tester can easily be failed by a small dust or a thin scratch, so it should be emphasized that very careful and precise work by a skilled person is indispensable. No other device can prove the accuracy except for a superior standard owned by a company authorized by Japan Calibration Service System (JCSS). At last, transducers are calibrated and the constants C and D are adjusted if necessary. Results of annual maintenance are shared between operator and inspectorate.

5.5 Replacement of transducer, longevity and diagnostics

The prototype ELTM system used for initial tank calibration and the first transfer of plutonium nitrate solution was duplicated and installed independently for each tank in 1984. Automated functions were added, but the full scale of ΔP_L transducer was still remained as 15 psi or 103 kPa which was different from the current one as 6 psi or 41 kPa. Since then, eight pressure transducers for four tanks have been continuously running in parallel. History of the replacements of transducers is shown in Table 2. The transducers used for ΔP_L were replaced by the transducers used for ΔP_D and the current model was employed for ΔP_D in 1993. Thus, four transducers have been continuously used initially for ΔP_D and later for ΔP_L since 1984. Another four transducers have been continuously used for ΔP_D since 1993.

We have experienced unexpected replacement of the transducer twice, and planned replacement once. The first one was unexpected in 2000, which was 7 years after the employment. The second one was unexpected in 2008, which was 15 years after the employment. The third one was planned in 2008, which was 24 years after the employment. Other five transducers are now running and their performance is normal over 15 years. According to the knowledge by the manufacturer, longevity of the transducer was estimated to be around ten years. Actual longevity was over 15 years except for the transducer failed in 2000. Static measurement environment compared to the general environment supposed by the manufacturer may prolong the longevity.

Table 2 History of transducer's replacements

Purpose		1984- (Prototype duplicated)	1993 - (Demonstrated)		2005 - (Improved)	
Tank #1	ΔP_L	215D 15 psi	5006D 6 psi	→	5006D 6 psi	5306D 6 psi *2
	ΔP_D	5006D 6 psi	5206D 6 psi	→	5206D 6 psi	5306D 6 psi *2
Tank #2	ΔP_L	215D 15 psi	5006D 6 psi	→	5006D 6 psi	5006D 6 psi
	ΔP_D	5006D 6 psi	5206D 6 psi	→	5206D 6 psi	5206D 6 psi
Tank #3	ΔP_L	215D 15 psi	5006D 6 psi	→	5006D 6 psi	5006D 6 psi
	ΔP_D	5006D 6 psi	5206D 6 psi	→	5206D 6 psi	5206D 6 psi
Tank #4	ΔP_L	215D 15 psi	5006D 6 psi	→	5006D 6 psi	5006D 6 psi
	ΔP_D	5006D 6 psi	5206D 6 psi	5306D 6 psi *1	5306D 6 psi	5306D 6 psi

*1 Replaced due to failure in 2000 *2 Replaced due to aging in 2008

Typical symptom of the failure due to aging is occurrence of instability observed unexpectedly at calibration or zero adjustment. Time available for use is very short after the occurrence, thus it is necessary to replace to the new one immediately. After the unexpected replacement in 2008, historical trend of the result of monthly calibration for the transducer was investigated. As a result, a characteristic behavior showing a sign of failure due to aging in advance of the occurrence of instability was found. The behavior was a rapid shift in T_0 which was resonant frequency at zero pressure shown in Figure 13, where T_0 was changed in 2000 as a result of replacement and T_0 was changed in 2008 as a result of aging. After this finding, historical trends of monthly calibrations of all transducers were investigated, then a weak behavior was found for the 24 years old transducer. Replacement was immediately planned and carried out, although no symptom of instability had been observed. Therefore, we found a useful diagnostic tool for replacement. Monitoring of T_0 will be continued in every calibration.

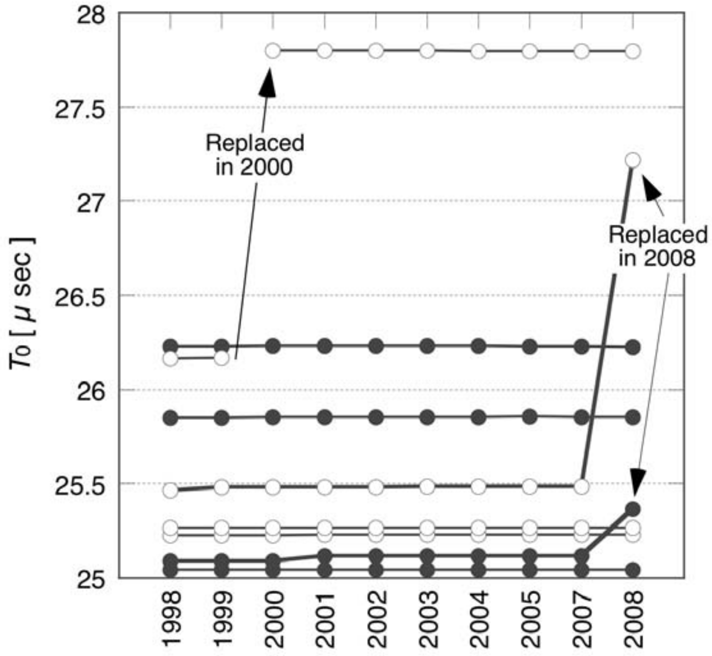


Figure 13 Behavior of T_0 for each transducer showing a sign of failure due to aging

5.6 Improvements

Based on the experience of replacement in 2000, layout of pipes and components of ELTM were re-designed and re-constructed at the end of 2004 to improve accessibility to the transducer. Conducting pipes were arranged not to disturb the replacement work and components were categorized into several replaceable units. As a result, the time necessary for replacement reduced from a couple of days to one hour. Two important advantages were obtained : 1) Preventive maintenance which consume cost and increase waste became not necessary because time of interruption of measurement became be permissible ; 2) Obtaining longevity data became possible because transducer could be used until the end of its lifetime. These improvements demonstrated their ability in the second and the third replacement in 2008. In addition, some improvements of software to ensure data treatment at power failure or to generate output more suitable to make a trend chart were added.

Meanwhile, the air put into dip-tube is well humidified but it normally yields no sweat inside the conducting pipe as long as the atmospheric temperature of the work area is well controlled. For this reason, a copper pipe suitable to obtain good sealing performance at ease has been used for the conducting pipe. However, we found a green rust at the time of annual maintenance. Estimated reason was a rapid variation of the atmospheric temperature of the work area caused unexpectedly at the time of periodical maintenance of air conditioning system. Therefore, we exchanged the material to stainless steel though it requires some attentions to obtain good sealing performance.

Combination of T_0 monitoring and quick replacement technique enabled an effective subsequent maintenance instead of preventive maintenance. Observation of the behavior related to aging will be continued and the results will be accumulated.

5.7 Update of certifications to ensure traceability

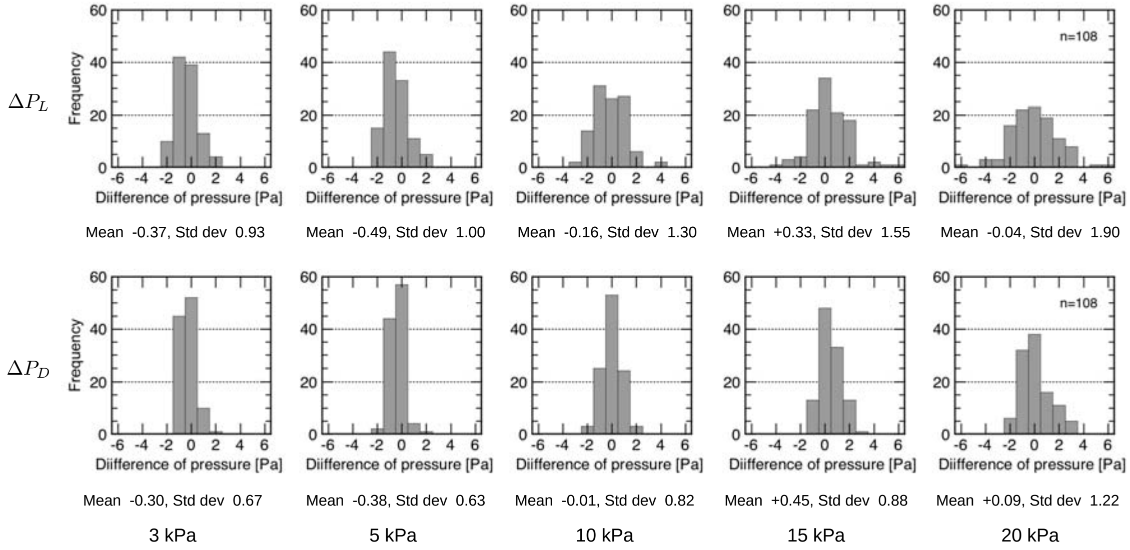
Certifications giving cross-section area of the cylinder at certain temperature and mass of disks have to be periodically updated at a frequency required by JCSS technical guide JCT 20501 published by National Institute of Technology and Evaluation (NITE). For this purpose, the dead-weight tester is brought out from the facility and carried in to a company authorized by JCSS, then compared with a superior standard which is traceable to the standard at National Metrology Institute of Japan (NMIJ). The current frequency is every three years. The results are provided to inspectorate if required. Expansion of cross-section area due to wearing has not been observed.

6. Volume measurement uncertainty proved by demonstration

Actual pressure measurement uncertainty practically achieved and sustained during demonstration was derived statistically from a number of calibrations of pressure transducers used for input tank. Then, actual volume measurement uncertainty was evaluated based on the uncertainty propagation same as Figure 2. Another comprehensive measurement uncertainty including sampling and other error source(s) was obtained statistically from the comparison test between measured density by ELTM and analyzed density by VTDM. In addition, a possibility of long-term change in gap length and level-volume relationship was evaluated from the results of input tank re-calibrations. At last, actual volume measurement uncertainty including knowledge obtained by demonstration was evaluated both for type A and for type B uncertainty, then a new target value for volume measurement uncertainties by dip-tube to be used for nuclear material accountancy was proposed.

6.1 Calibrations of differential pressure transducers

Frequency distribution of the difference between reference and transducer is shown in Figure 14 at each reference pressure (3, 5, 10, 15, 20 kPa). The distributions were obtained from 108 calibration records since 1989. One of the transducer was changed in 1993 as shown in Table 2, so the population is not focused on characteristics of each transducer but focused on the measurement system. Profile of distribution is a single peak in every case.



Mean -0.37, Std dev 0.93 | Mean -0.49, Std dev 1.00 | Mean -0.16, Std dev 1.30 | Mean +0.33, Std dev 1.55 | Mean -0.04, Std dev 1.90

Mean -0.30, Std dev 0.67 | Mean -0.38, Std dev 0.63 | Mean -0.01, Std dev 0.82 | Mean +0.45, Std dev 0.88 | Mean +0.09, Std dev 1.22

3 kPa | 5 kPa | 10 kPa | 15 kPa | 20 kPa

Figure 14 Frequency distributions of calibration results (measured - reference)

The distribution of ΔP_D at 3 kPa was less than 0.5 ± 0.7 Pa. The standard deviation becomes larger at higher pressure, but it does not affect pressure measurement. The distributions of ΔP_L at 15 - 20 kPa were less than 0.5 ± 1.9 Pa. These standard deviations consist of repeatability of the transducer, fluctuation of the reference, and effect of deviation of zero point and/or variation of temperature sensitivity. Another random component is pressure oscillation around 1 Pa. Therefore, actual random component of pressure measurement uncertainty for oscillating wave is calculated as square-root of sum of squares, which is 1.3 Pa ($= \sqrt{0.7^2 + 1^2}$) and 2.2 Pa ($= \sqrt{1.9^2 + 1^2}$) for ΔP_D and ΔP_L , respectively.

There is a trend for both ΔP_L and ΔP_D that the average is most negative at 5 kPa and most positive at 15 kPa. However, the center and the width of the trend is close to zero and less than 0.5 Pa respectively. The estimated reason is an approximation error of the equation to convert frequency to pressure, or a residue after optimization of the constants C and D. This trend is very similar to the one at initial setting, so it is acceptable.

6.2 Actual propagation of measurement uncertainty

Actual propagation of measurement uncertainty corresponds to Figure 2 was evaluated and shown in Figure 15. Random measurement uncertainties for plutonium nitrate solution were 1.3 Pa and 2.2 Pa, thus relative uncertainties were 0.044 % of 3 kPa and 0.013 % of 17 kPa for ΔP_D and ΔP_L , respectively. Random measurement uncertainties at initial tank calibration carried out in 1982 by prototype ELTM were 1.4 Pa ($= \sqrt{1^2 + 1^2}$) and 2.2 Pa ($= \sqrt{2^2 + 1^2}$) including pressure oscillation, thus relative uncertainties were 0.070 % of 2 kPa and 0.019 % of 12 kPa for ΔP_D and ΔP_L , respectively. Measurement uncertainty of reference liquid weight was 0.01% because standard deviation was 0.01 kg for the full scale 100 kg. Uncertainty of reference liquid density (ion exchanged water) was 0.013% supposing temperature measurement uncertainty as 0.5 degree. Fitting error was added as 0.013 % (0.04 L for 300 L).

Evaluated random measurement uncertainty of density for plutonium nitrate solution was 0.084 % as one sigma. Evaluated random measurement uncertainty of volume for plutonium nitrate solution was 0.091 % as one sigma. They are type A uncertainties obtained from statistics above. Type B uncertainty is composed of a deviation of the average between reference and transducer and an uncertainty of measurement bias correction mentioned in the section 3.6. The former is less than 0.017 % as 0.5 Pa of 3 kPa and the latter is less than 0.052 %. The sum is less than 0.055 %. Deviation of the average at tank initial calibration had already been corrected. They are the measurement uncertainties related to ELTM practically achieved, sustained and proved by demonstration over fifteen years.

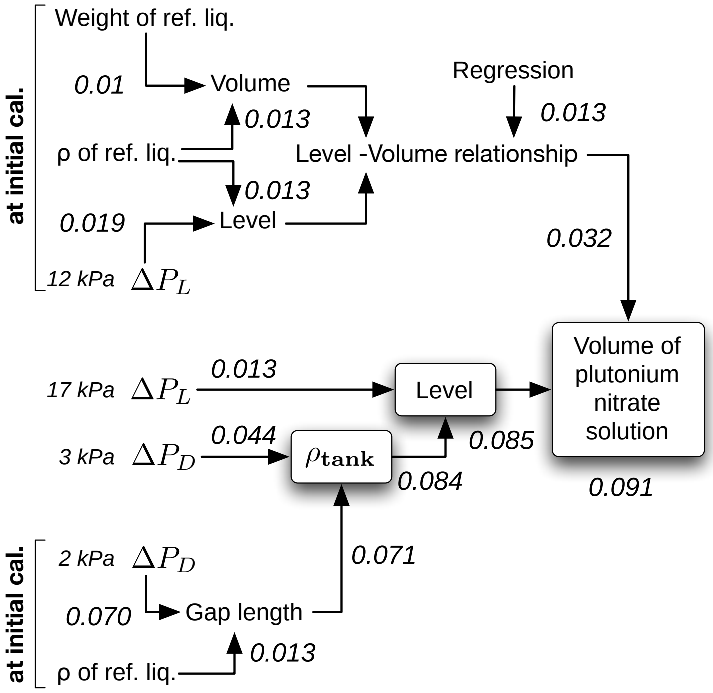


Figure 15 Actual propagation of measurement uncertainty (% of measured)

6.3 Comparison tests between measured density and analyzed density

There are two solution densities measured by ELTM and analyzed by VTDM at different temperature as shown in Figure 1. Frequency distribution of the difference between two solution densities after temperature adjustment is shown in Figure 16, which was obtained from 49 comparison data since 1994. The difference (ELTM - VTDM) was 0.0005 ± 0.002 $g \cdot cm^{-3}$. The mean was very small which was equivalent to 0.03% of solution density around 1.45 $g \cdot cm^{-3}$. The mean includes theoretically the bias due to thermal expansion of gap length which is 0.0017 % per degree (refer to section 3.3) and temperature difference around 16 °C (between initial calibration and measurement). As for the bubble effect and tank thermal expansion effect (refer to section 3.4), ΔP_D measurement is free from the bubble effect, so the effect of thermal expansion of gap length remains without cancellation. The mean was consistent with the magnitude and direction of the bias, and the residue was smaller than the uncertainty of other bias corrections.

This consistency also proves that a bias in regard to condensation of solution by sampling system, sampling operation or other error source(s) is practically very small though it has been considered to be possible. This is an important finding obtained from the demonstration regarding the circulating sampling system and its condition.

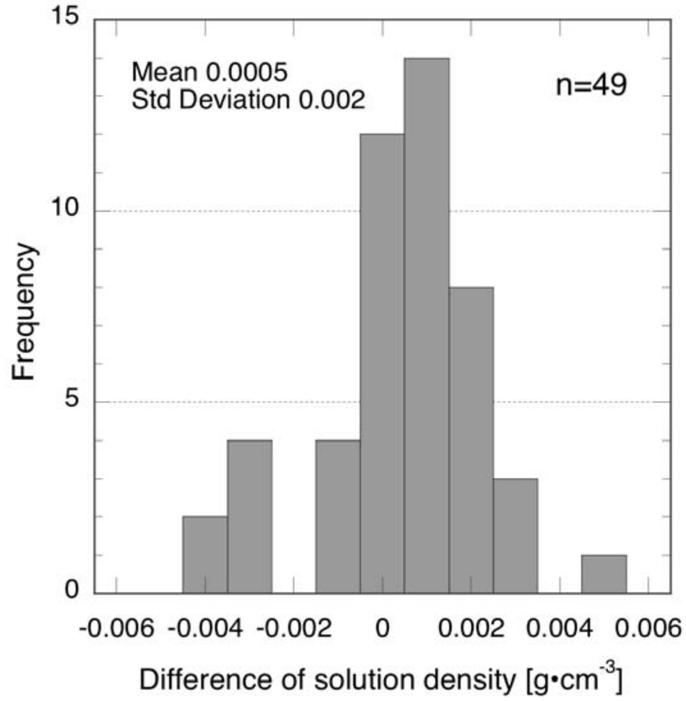


Figure 16 Frequency distribution of comparison test results

On the other hand, the standard deviation of the difference (0.002 g•cm^{-3}) was equivalent to 0.14 % of solution density, which was a little larger than expected. The expected was 0.11 % calculated from sum of squares of type A uncertainties correspond to ELTM (0.084 %), VTDM (0.05 %) and temperature measurement (0.04 % as 0.5 °C). In addition, the profile had a small second peak or a long tail which means some existence of uncontrolled and nonprobabilistic source(s) of measurement uncertainty. Estimated source was something like a small unstable precipitation occasionally yields and leaves at the tip of dip-tube. We have experienced half-clogging both in major tube and in minor tube, so it was probable that the effect of such precipitation appears positively and negatively in density measurement by ELTM.

Contribution of the uncontrolled and nonprobabilistic source(s) of measurement uncertainty on the comparison test of two densities could be estimated inversely as $0.09\% = \sqrt{0.14^2 - 0.084^2 - 0.05^2 - 0.04^2}$ which was equivalent to around 3 Pa of 3 kPa (ΔP_D measurement). The estimated source was considered to be probable, and the magnitude was as large as the type A uncertainty of ELTM itself. Therefore, this is also an important finding obtained from demonstration regarding effect of precipitation and stability of gap length.

6.4 Re-calibration of accountability tank

Re-calibration of input accountability tank has been carried out using nitric acid (~ 0.5 N) as reference solution every year (except for special cases agreed with inspectorate) to confirm consistency of gap length and level-volume relationship. Two comparison tests are performed at re-calibration as shown in Figure 17 where acceptable range is evaluated as one sigma of the uncertainty for each test.

Test 1 is for the difference between measured volume and total fed volume added to heel volume. Measured volume is calculated from reference density, ΔP_L and current level-volume relationship. Fed volume is calculated from reference density and fed weight. The purpose is to confirm current level-volume relationship. Test 2 is for the difference between measured density and density of nitric acid. Measured density is calculated from ΔP_D and current gap-length. The purpose is to confirm current gap-length. Uncertainty of the reference liquid density is set at 0.05 % considering measurement uncertainty of VTDM and difficulty of complete clean out before re-calibration. As a result, acceptable range of the comparison tests are evaluated as 0.08 % and as 0.11 % for the difference of volume and for the difference of density, respectively.

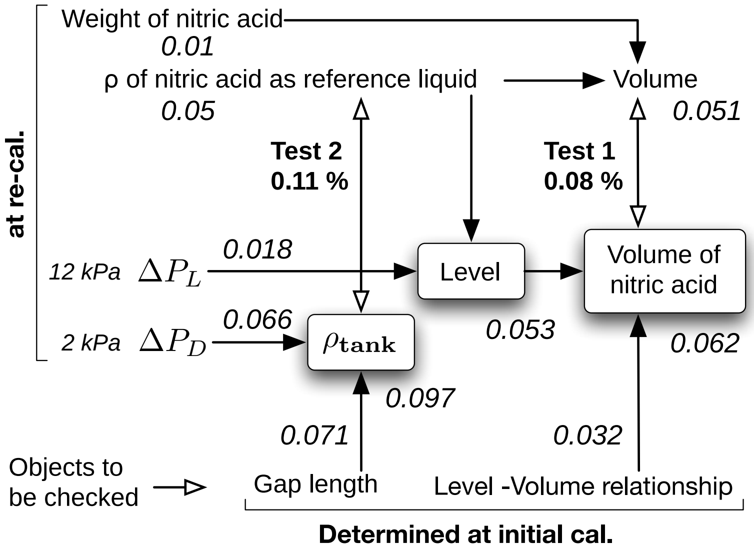


Figure 17 Propagation of meas. uncertainty at tank re-calibration (% of measured)

As a result of re-calibrations during demonstration, difference of density (Test 2) has been within the acceptable range and no trend has been observed. However a small trend concerning the difference of volume (Test 1) has been observed within the acceptable range. The trend was composed of fitting error at initial calibration and linear slope proportional to liquid level. An example is shown in Figure 18.

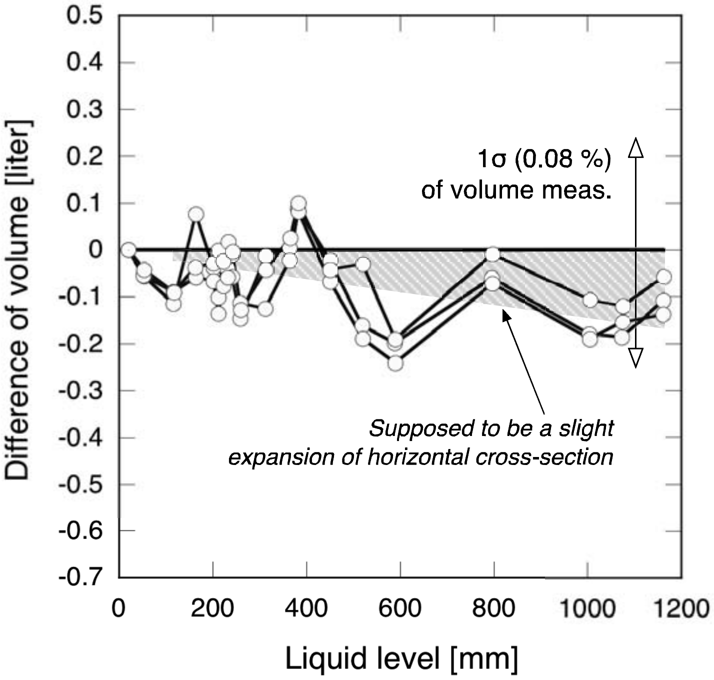


Figure 18 Recent example of volume differences at tank re-calibration (calculated from equation - fed)

An averaged gradient of the slope throughout all re-calibrations was a little negative estimated to be around 0.05 %, but the gradient fluctuates widely in each year. The small negative linear slope could be an expansion of horizontal cross-section due to corrosion developed very slowly at the temperature of plutonium nitrate solution. We evaluated the corrosion thickness to be 10 - 30 μm from the negative slope. Such slow flat corrosion is considered to be unavoidable but difficult to be confirmed quantitatively because there is very little corrosion data at low temperature. Our rough evaluation was consistent with the report by Takeda (1995)(28) where corrosion test was performed in plutonium nitrate solution of 8 N, 290 gPu/liter at 313 K and the result was around 0.01-0.02 $g \cdot m^{-2} \cdot hr$.

As a result, effect of unavoidable change in level-volume relationship due to corrosion has not been detected beyond the measurement uncertainty, but the order was estimated roughly around 0.05 % from the result of all re-calibrations. This is also an important finding obtained from demonstration regarding stability of level-volume relationship. In addition, an appropriate or recommended frequency of re-calibration or new initial calibration is once every five years or less from the technical point of view. Another method is strongly desired for the purpose of safeguards to verify periodically the level-volume relationship and the gap length, because it yields a lot of waste liquid and disturbs process operation.

6.5 Proposal of a new target value

Based on the results of section 3.6, section 4.6 and this section, it became clear that volume measurement uncertainties by dip-tube for plutonium nitrate solution consists of : 1) less than 0.052 % as an uncertainty related to bias corrections ; 2) less than 0.017 % as an uncertainty related to discrepancy between reference and transducer after optimization ; 3) 0.091 % as random component of practical measurement uncertainty by ELTM ; 4) roughly less than 0.05 % as long-term change in level-volume relationship corresponds to expansion of horizontal cross-section possibly due to corrosion ; 5) around 0.09 % as uncontrolled but probable random component possibly caused by a small unstable precipitation occasionally yields and leaves at the tip of dip-tube. The first one, the second one and the fourth one belong to type B uncertainty of systematic character, and their combination is less than 0.074 %. The third one and the fifth one belong to type A uncertainty as random component, and their combination is 0.13 %. The fourth one and the fifth one are quite new findings as a result of demonstration over fifteen years and cumulative plutonium throughput is six tons. The third one is the proof of appropriate maintenance during demonstration.

In conclusion, we would like to propose a new set of target values applied to volume measurement uncertainties by dip-tube to be used for nuclear material accountancy based on accumulated data during the demonstration. The values are less than 0.1 % for component of systematic character and 0.15 % for random component. These uncertainties include effects caused by long term use of the accountability tank such as a small unstable precipitation occasionally yields and leaves at the tip of dip-tube and/or flat corrosion developed very slowly at the temperature of plutonium nitrate solution. The obtained uncertainties were comparable to the analytical measurements used for plutonium accounting.

7. Conclusions

ELTM system for accurate volume measurement of plutonium nitrate solution to determine plutonium weight transferred from TRP to PCDF has been designed carefully, put into practice by operator and by inspectorate, developed continuously and demonstrated successfully since 1982. The system is composed of a set of digiquartz® differential pressure transducers, a dead-weight tester as a reference, sequential valve control to adjust zero point and calibration, pneumatic damping of pressure oscillation. Measurement biases have been properly evaluated and corrected since the first practical apply to the initial tank calibration in 1982. As a result of statistical evaluation of the accumulated data regarding to transducer calibrations, tank calibrations and comparison tests of densities between the system and analytical method during the demonstration over fifteen years, measurement uncertainties practically achieved and kept were confirmed to be the half of the current target uncertainties admitted internationally and comparable to the analytical measurements. Expected performance of the system in design base has been kept and proved by daily check, monthly and occasional calibration, and annual maintenance. Reference pressures by dead-weight tester was periodically ensured to be traceable to JCSS. Effects by long term use of the tank where cumulative plutonium throughput was 6 tons were found though they were less than 0.1 %. Half-clogging of dip-tube and unexpected replacement of transducer were experienced and resolved.

- Pressure measurement uncertainty practically achieved and maintained were evaluated to be less than 0.5 ± 0.7 Pa for ΔP_D and less than 0.5 ± 1.9 Pa for ΔP_L as one sigma which includes repeatability of the transducer, repeatability of the reference pressure and effect of temperature variation.
- Volume measurement uncertainty as random component practically achieved and maintained were evaluated to be less than 0.91 % as one sigma which includes effect of pressure oscillation and measurement uncertainties at initial tank calibration such as weight and density measurement of reference liquid.
- Volume measurement uncertainty of systematic character is composed of uncertainties related to bias correction as 0.052 % and uncertainty related to discrepancy between reference and transducer after optimization as 0.017 %.
- There are five major measurement biases : a) thermal expansion of solution; b) air column effect ; c) thermal expansion of tank ; d) bubble effect ; and e) pressure loss. For a) and b), they had already been corrected since the initial tank calibration. For c) and d), they were evaluated and proved not negligible but over half was cancelled within current operational conditions. Corrections will be introduced at the time of high-burn up plutonium reprocessing in a next few years. For pressure loss, correction is not necessary in our facility.

- As a result of comparison tests, difference between measured density by ELTM and analytically determined density was less than 0.0005 ± 0.002 $g \cdot cm^{-3}$ as one sigma, though they were obtained from quite different sources. Measurement bias in regard to sampling was evaluated to be small.
- Effects by long term use of the tank were not detected statistically but a small trend was found where cumulative plutonium throughput was 6 tons : 1) roughly less than 0.05 % as long-term change in level-volume relationship corresponds to expansion of horizontal cross-section possibly due to corrosion ; 2) around 0.09 % as uncontrolled but probable random component possibly caused by a small unstable precipitation occasionally yields and leaves at the tip of dip-tube.
- Total volume measurement uncertainties including systematic characters and the effect of log-time use were evaluated less than 0.074 % (systematic character) and 0.13 % (random) as one sigma, which was comparable to the analytical measurements used for plutonium accounting.
- Half-clogging of dip-tube were detected by careful monitoring and treated timely, however it was difficult to avoid recurrence.
- Calibrations and maintenances accompanied by opening conducting pipes were done safely owing to the automated valve sequential control.
- We have experienced unexpected replacement of the transducer twice, and planned replacement once. Longevity of digiquartz® transducer was evaluated to be longer than 15 years.
- Improvement of accessibility to the transducer at the replacement was effective : a) time for replacement was reduced to one hour ; b) preventive maintenance was no longer necessary ; and c) obtaining longevity data became be possible.
- Monitoring of T_0 was useful to diagnose appropriate timing of replacement of the transducer.

Finally, a new set of target values applied to volume measurement uncertainties by dip-tube to be used for nuclear material accountancy was proposed based on accumulated data during the demonstration . The values are less than 0.1 % for component of systematic character and 0.15 % for random component. These uncertainties include effects by long term use of the accountability tank. Volume measurement is no longer the largest source of measurement uncertainty for determination of plutonium weight in a tank.

Acknowledgment

This work was completed by steady and consistent thinkings and efforts without fail by PCDF operators, members of analytical section and management section, manufacturer and inspectorates. I wish to acknowledge especially to Katsuya KOISO and Hiroyuki HAYASHI for absolutely perfect operation and maintenance, Yasunobu MUKAI, Hironobu NAKAMURA and Hideki TANAKA for constructive suggestions and cooperation, Toshihide KABUKI for helpful frank communication with inspectorate, Dr. Tsunemichi KAWA for technical suggestions of precise measurement and Prof. Masanori ARITOMI for academic approach of dissertation.

References

(1) O. Yamamura, R. Yamamoto, S. Nomura, Y. Fujii : "Development of safeguards and maintenance technology in Tokai Reprocessing Plant," Progress in Nuclear Energy, 50, pp. 666-673 (2008).

(2) H. Oshima, T. Tamura, M. Koizumi : "Outline of the Coconversion Facility of Pu-U Nitrate Solution to the Mixed-Oxide Powder Using a Microwave Heating Method," Transaction of American Nuclear Society (ANS), vol. 40 (1982).

(3) H. Oshima : "Development of Microwave Heating Method for Co-Conversion of Plutonium-Uranium Nitrate to MOX Powder," J. of Nuclear Science Technology, 26 (1), pp. 161-166 (1989).

(4) Y. Kato, T. Kurita, T. Abe : "Dielectric Properties of Uranium and Plutonium Nitrate Solution and the Oxide Compounds Formed in the De-nitration Process by the Microwave Heating Method," J. of Nuclear Science Technology, 41 (8), pp. 857-862 (2004).

(5) T. Hosoma, K. Ichige, Y. Takahashi : "History of 20 years Demonstration of Pu-U Co-Conversion Process Using a Microwave Heating Direct Denitration Method," No. 24, pp. 11-26 (2004) (in Japanese).

(6) H. Aigner, R. Binner, E. Kuhn, et. al. : "International Target Values 2000 for Measurement Uncertainties in Safeguarding Nuclear Materials," ESARDA (European Safeguards Research and Development Association) BULLETIN, No. 31, pp.39-68 (2002).

(7) D. Reilly, N. Ensslin, H. Smith Jr., S. Kreiner : "Passive Nondestructive Assay of Nuclear Materials (PANDA)," NUREG/CR--5550 (1991) and PANDA 2007 Addendum.

(8) O. Yamamura, R. Yamamoto, S. Nomura : "Historical Role of the Tokai Reprocessing Plant in the Establishment of Safeguards Technologies," J. of Nuclear Materials Management, vol. 36, 3, pp. 36-45 (2008).

(9) H. O. Menlove, T. D. Reilly : "The Development and Implementation of NDA Equipment at the IAEA," Proc. INMM (2007).

(10) Y. Noguchi, N. Majima, H. Nakamura, T. Iwamoto : "Material Accountancy and NDA approach for U-Pu Co-Denitration Area at Rokkasyo Reprocessing Plant (RRP)," Proc. INMM (2005).

(11) Y. Noguchi, H. Nakamura, H. Nakayashiki, T. Iwamoto : "Development and Calibration of Temporary Canister Verification System (TCVS) to assay Pu Inventory in Intermediate MOX Canisters at RRP," Proc. INMM (2007).

(12) H. Nakamura, A. Takaya, T. Iwamoto, et. al. : "First trial to study the feasibility of direct plutonium mass measurement in a process tank by a new NDA," Proc. INMM (2009).

(13) F. E. Jones, G. L. Harris : "ITS-90 Density of Water Formulation for Volumetric Standards Calibration," J. of Research of the National Institute of Standards and Technology, vol. 97, 3, pp. 335-340 (1992).

(14) S. Sakurai : "Modified Density Equation for Aqueous Solution with Plutonium(IV) and Nitric Acid," JAERI-M 91-117 (1991).

(15) A. Maimoni : “Density and Radiolytic Decomposition of Plutonium Nitrate Solutions,” UCRL-52727 (1979).

(16) S. Kumar, S. B. Koganti : “Prediction of Densities of Mixed Aqueous Solutions of Electrolytes - $UO_2(NO_3)_2$, $Pu(NO_3)_4$ and Nitric Acid,” J. of Nuclear Science Technology, 34(4), pp. 410-412 (1997).

(17) N. Charrin, Ph. Moisy, P. Blanc : “Contribution of the concept of simple solutions to calculation of the density of ternary and quaternary solutions of thorium(IV) or plutonium(IV) nitrate : $An(NO_3)_4/UO_2(NO_3)_2/HNO_3/H_2O$,” Radiochim. Acta 88, 445-451 (2000).

(18) T. Hosoma, Y. Mukai : “Reduction of Fluctuation and Small Bias Observed in Continuous Volume Monitoring taken in an Annular Tank for Plutonium Nitrate,” Proc. IAEA Symposium (2006).

(19) “Nuclear fuel technology - Tank calibration and volume determination for nuclear materials accountancy - ,” Part 1 to Part 6 (2007 to 2009)

(20) H. Higuchi, S. Takeda, S. Uchikoshi, et. al. : “High Quality Tank Calibration Study,” Proc. IAEA Symposium (2001).

(21) T. Hosoma, M. Aritomi, T. Kawa : “Numerical Calculation and Measurement of Bubble Shape and Excess Pressure in Dip-Tube Pressure Measurement,” Nuclear Technology, vol. 120, pp. 121-135 (1997).

(22) F. Bashforth and J. C. Adams : “An attempt to test the theories of capillary action,” Cambridge University Press (1883).

(23) S. Hartland, R. W. Hartley : “Axisymmetric Fluid-Liquid Interfaces,” Elsevier (1976).

(24) T. Hosoma, M. Aritomi, T. Kawa : “Formulae to Correct Excess Pressure and Pressure Shift to be used in Volume Measurement for Plutonium Nitrate Solution,” Nuclear Technology, vol. 129, pp. 218-235 (2000).

(25) Paroscientific Inc. : “Differential Pressure Transducers Series 5300,” available <http://www.paroscientific.com/pdf/5300.pdf> (accessed 2010-4-30)

(26) Tsukasa Sokken Co. Ltd. “Ultra precision Digiquarts manometers,” available <http://www.sokken.co.jp/en/index.html> (accessed 2010-4-30)

(27) DH-Budenberg Inc. : “Portable Dead-Weight Testers,” available <http://www.dh-budenberginc.com/media/techImage125.PDF> (accessed 2010-4-30)

(28) S. Takeda, T. Nagai, S. Yasu, T. Koizumi : “Corrosion Performance of Several Metals in Plutonium Nitrate Solution,” Zairyo-to-Kankyo, 44, pp.24-29 (1995) (in Japanese).

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国際単位系（SI）

表１．SI 基本単位

基本量	SI 基本単位	
	名称	記号
長さ	メートル	m
質量	キログラム	kg
時間	秒	s
電流	アンペア	A
熱力学温度	ケルビン	K
物質量	モル	mol
光度	カンデラ	cd

表２．基本単位を用いて表されるSI組立単位の例

組立量	SI 基本単位	
	名称	記号
面積	平方メートル	m^{2}
体積	立法メートル	m^{3}
速さ，速度	メートル毎秒	m/s
加速度	メートル毎秒毎秒	m/s^{2}
波数	毎メートル	m^{-1}
密度，質量密度	キログラム毎立方メートル	kg/m^{3}
面積密度	キログラム毎平方メートル	kg/m^{2}
比体積	立方メートル毎キログラム	m^{3}/kg
電流密度	アンペア毎平方メートル	A/m^{2}
磁界の強さ	アンペア毎メートル	A/m
量濃度(a)，濃度	モル毎立方メートル	mol/m^{3}
質量濃度	キログラム毎立法メートル	kg/m^{3}
輝度	カンデラ毎平方メートル	cd/m^{2}
屈折率(b)	（数字の）　1	1
比透磁率(b)	（数字の）　1	1

(a)　量濃度（amount concentration）は臨床化学の分野では物質濃度（substance concentration）ともよばれる。

(b)　これらは無次元量あるいは次元１をもつ量であるが、そのことを表す単位記号である数字の１は通常は表記しない。

表３．固有の名称と記号で表されるSI組立単位

組立量	SI 組立単位			
	名称	記号	他のSI単位による表し方	SI基本単位による表し方
平面角	ラジアン(b)	rad	1 (b)	m/m
立体角	ステラジアン(b)	sr(c)	1 (b)	m^{2}/m^{2}
周波数	ヘルツ(d)	Hz		s^{-1}
力	ニュートン	N		m kg s^{-2}
圧力，応力	パスカル	Pa	N/m^{2}	m^{-1} kg s^{-2}
エネルギー，仕事，熱量	ジュール	J	N m	m^{2} kg s^{-2}
仕事率，工率，放射束	ワット	W	J/s	m^{2} kg s^{-3}
電荷，電気量	クーロン	C		s A
電位差（電圧），起電力	ボルト	V	W/A	m^{2} kg s^{-3} A^{-1}
静電容量	ファラド	F	C/V	m^{-2} kg^{-1} s^{4} A^{2}
電気抵抗	オーム	Ω	V/A	m^{2} kg s^{-3} A^{-2}
コンダクタンス	ジーメンス	S	A/V	m^{-2} kg^{-1} s^{3} A^{2}
磁束	ウエーバ	Wb	Vs	m^{2} kg s^{-2} A^{-1}
磁束密度	テスラ	T	Wb/m^{2}	kg s^{-2} A^{-1}
インダクタンス	ヘンリー	H	Wb/A	m^{2} kg s^{-2} A^{-2}
セルシウス温度	セルシウス度(e)	℃		K
光束	ルーメン	lm	cd sr(c)	cd
照度	ルクス	lx	lm/m^{2}	m^{-2} cd
放射性核種の放射能(f)	ベクレル(d)	Bq		s^{-1}
吸収線量, 比エネルギー分与, カーマ	グレイ	Gy	J/kg	m^{2} s^{-2}
線量当量, 周辺線量当量, 方向性線量当量, 個人線量当量	シーベルト(g)	Sv	J/kg	m^{2} s^{-2}
酸素活性	カタール	kat		s^{-1} mol

(a)SI接頭語は固有の名称と記号を持つ組立単位と組み合わせても使用できる。しかし接頭語を付した単位はもはやコヒーレントではない。

(b)ラジアンとステラジアンは数字の１に対する単位の特別な名称で、量についての情報をつたえるために使われる。実際には、使用する時には記号rad及びsrが用いられるが、習慣として組立単位としての記号である数字の１は明示されない。

(c)測光学ではステラジアンという名称と記号srを単位の表し方の中に、そのまま維持している。

(d)ヘルツは周期現象についてのみ、ベクレルは放射性核種の統計的過程についてのみ使用される。

(e)セルシウス度はケルビンの特別な名称で、セルシウス温度を表すために使用される。セルシウス度とケルビンの単位の大きさは同一である。したがって、温度差や温度間隔を表す数値はどちらの単位で表しても同じである。

(f)放射性核種の放射能（activity referred to a radionuclide）は、しばしば誤った用語で”radioactivity”と記される。

(g)単位シーベルト（PV,2002,70,205）についてはCIPM勧告2（CI-2002）を参照。

表４．単位の中に固有の名称と記号を含むSI組立単位の例

組立量	SI 組立単位		
	名称	記号	SI 基本単位による表し方
粘度	パスカル秒	Pa s	m^{-1} kg s^{-1}
力のモーメント	ニュートンメートル	N m	m^{2} kg s^{-2}
表面張力	ニュートン毎メートル	N/m	kg s^{-2}
角速度	ラジアン毎秒	rad/s	m m^{-1} s^{-1}=s^{-1}
角加速度	ラジアン毎秒毎秒	rad/s^{2}	m m^{-1} s^{-2}=s^{-2}
熱流密度，放射照度	ワット毎平方メートル	W/m^{2}	kg s^{-3}
熱容量，エントロピー	ジュール毎ケルビン	J/K	m^{2} kg s^{-2} K^{-1}
比熱容量，比エントロピー	ジュール毎キログラム毎ケルビン	J/(kg K)	m^{2} s^{-2} K^{-1}
比エネルギー	ジュール毎キログラム	J/kg	m^{2} s^{-2}
熱伝導率	ワット毎メートル毎ケルビン	W/(m K)	m kg s^{-3} K^{-1}
体積エネルギー	ジュール毎立方メートル	J/m^{3}	m^{-1} kg s^{-2}
電界の強さ	ボルト毎メートル	V/m	m kg s^{-3} A^{-1}
電荷密度	クーロン毎立方メートル	C/m^{3}	m^{-3} sA
表面電荷	クーロン毎平方メートル	C/m^{2}	m^{-2} sA
電束密度，電気変位	クーロン毎平方メートル	C/m^{2}	m^{-2} sA
誘電率	ファラド毎メートル	F/m	m^{-3} kg^{-1} s^{4} A^{2}
透磁率	ヘンリー毎メートル	H/m	m kg s^{-2} A^{-2}
モルエネルギー	ジュール毎モル	J/mol	m^{2} kg s^{-2} mol^{-1}
モルエントロピー, モル熱容量	ジュール毎モル毎ケルビン	J/(mol K)	m^{2} kg s^{-2} K^{-1} mol^{-1}
照射線量（Ｘ線及びγ線）	クーロン毎キログラム	C/kg	kg^{-1} sA
吸収線量率	グレイ毎秒	Gy/s	m^{2} s^{-3}
放射強度	ワット毎ステラジアン	W/sr	m^{4} m^{-2} kg s^{-3}=m^{2} kg s^{-3}
放射輝度	ワット毎平方メートル毎ステラジアン	W/(m^{2} sr)	m^{2} m^{-2} kg s^{-3}=kg s^{-3}
酵素活性濃度	カタール毎立方メートル	kat/m^{3}	m^{-3} s^{-1} mol

表５．SI 接頭語

乗数	接頭語	記号	乗数	接頭語	記号
10^{24}	ヨタ	Y	10^{-1}	デシ	d
10^{21}	ゼタ	Z	10^{-2}	センチ	c
10^{18}	エクサ	E	10^{-3}	ミリ	m
10^{15}	ペタ	P	10^{-6}	マイクロ	µ
10^{12}	テラ	T	10^{-9}	ナノ	n
10^{9}	ギガ	G	10^{-12}	ピコ	p
10^{6}	メガ	M	10^{-15}	フェムト	f
10^{3}	キロ	k	10^{-18}	アト	a
10^{2}	ヘクト	h	10^{-21}	ゼプト	z
10^{1}	デカ	da	10^{-24}	ヨクト	y

表６．SIに属さないが、SIと併用される単位

名称	記号	SI 単位による値
分	min	1 min=60s
時	h	1h =60 min=3600 s
日	d	1 d=24 h=86 400 s
度	°	1°=(π/180) rad
分	’	1’=(1/60)°=(π/10800) rad
秒	”	1”=(1/60)’=(π/648000) rad
ヘクタール	ha	1ha=1hm^{2}=10^{4}m^{2}
リットル	L，l	1L=11=1dm^{3}=10^{3}cm^{3}=10^{-3}m^{3}
トン	t	1t=10^{3} kg

表７．SIに属さないが、SIと併用される単位で、SI単位で表される数値が実験的に得られるもの

名称	記号	SI 単位で表される数値
電子ボルト	eV	1eV=1.602 176 53(14)×10^{-19}J
ダルトン	Da	1Da=1.660 538 86(28)×10^{-27}kg
統一原子質量単位	u	1u=1 Da
天文単位	ua	1ua=1.495 978 706 91(6)×10^{11}m

表８．SIに属さないが、SIと併用されるその他の単位

名称	記号	SI 単位で表される数値
バール	bar	1 bar=0.1MPa=100kPa=10^{5}Pa
水銀柱ミリメートル	mmHg	1mmHg=133.322Pa
オングストローム	Å	1 Å=0.1nm=100pm=10^{-10}m
海里	M	1 M=1852m
バーン	b	1 b=100fm^{2}=(10^{-12}cm)2=10^{-28}m^{2}
ノット	kn	1 kn=(1852/3600)m/s
ネーパ	Np	SI単位との数値的な関係は、対数量の定義に依存。
ベル	B	
デジベル	dB	

表９．固有の名称をもつCGS組立単位

名称	記号	SI 単位で表される数値
エルグ	erg	1 erg=10^{-7} J
ダイン	dyn	1 dyn=10^{-5}N
ポアズ	P	1 P=1 dyn s cm^{-2}=0.1Pa s
ストークス	St	1 St =1cm^{2} s^{-1}=10^{-4}m^{2} s^{-1}
スチルブ	sb	1 sb =1cd cm^{-2}=10^{4}cd m^{-2}
フォト	ph	1 ph=1cd sr cm^{-2} 10^{4}lx
ガル	Gal	1 Gal =1cm s^{-2}=10^{-2}ms^{-2}
マクスウェル	Mx	1 Mx = 1G cm^{2}=10^{-8}Wb
ガウス	G	1 G =1Mx cm^{-2} =10^{-4}T
エルステッド(c)	Oe	1 Oe ≙ (10^{3}/4π)A m^{-1}

(c)　３元系のCGS単位系とSIでは直接比較できないため、等号「 ≙ 」は対応関係を示すものである。

表10．SIに属さないその他の単位の例

名称	記号	SI 単位で表される数値
キュリー	Ci	1 Ci=3.7×10^{10}Bq
レントゲン	R	1 R = 2.58×10^{-4}C/kg
ラド	rad	1 rad=1cGy=10^{-2}Gy
レム	rem	1 rem=1 cSv=10^{-2}Sv
ガンマ	γ	1 γ =1 nT=10-9T
フェルミ		1フェルミ=1 fm=10-15m
メートル系カラット		1メートル系カラット = 200 mg = 2×10-4kg
トル	Torr	1 Torr = (101 325/760) Pa
標準大気圧	atm	1 atm = 101 325 Pa
カロリー	cal	1cal=4.1858J（「15℃」カロリー），4.1868J（「IT」カロリー）4.184J（「熱化学」カロリー）
ミクロン	µ	1 µ =1µm=10^{-6}m

（第8版，2006年改訂）