Proceedings of Metrologie ’99 Conference, France, 1999.

Intercomparison Tests of the NRLM Transfer Standard with the Primary Standards of NIST, BNM-LNE, OFMET and PTB for Small Mass Flow Rates of Nitrogen Gas

Shin-ichi Nakao, NRLM, Japan
John Wright, NIST, USA
Jean Barbe, BNM-LNE, France
Bernhard Niederhauser, Manuela Quintilii, OFMET, Switzerland
Dorothea Knopf, PTB, Germany

Abstract

The NRLM transfer standard was tested at the following national laboratories from April 1996 to June 1998 and compared with their primary standards over the flow range from 0.01 g/min to 100 g/min of nitrogen gas: the National Institute of Standards and Technology (NIST), the Swiss Federal Office of Metrology (OFMET), Bureau National de Metrologie - Laboratoire National D’essais (BNM-LNE) and Physikalisch-Technische Bundesanstalt (PTB). The results show good agreement for all of the laboratories over the flow range tested. A detailed examination of the data shows that the results between 0.2 g/min and 10 g/min agree within 0.2 %. Outside of this flow range, the variation of the results is greater than 0.2 % but smaller than 0.3 %. In the flow range near 1 g/min, the results agree within 0.1 %.

L'étalon de transfert du National Research Laboratory of Metrology (NRLM) a circulé dans des laboratoires nationaux entre avril 1996 et juin 1998 et comparé à leurs étalons primaires dans le domaine de 0,01 g/min à 100 g/min d'azote, savoir le National Institute of Standards and Technology (NIST), le Swiss Federal Office of Metrology (OFMET), le Bureau National de Mérologie - Laboratoire National d'Essais (BNM - LNE) et le Physikalisch Technische Bundesanstalt (PTB). Dans l'ensemble, les résultats montrent un accord meilleur que 0,3 percent sur tout le domaine étudié. Un examen approfondi des données montre que les résultats des comparaisons sont meilleurs dans le domaine de 0,2 g/min à 10 g/min où la dispersion des données est d'environ 0,2 percent. Dans le domaine de 1 g/min, la dispersion des valeurs est d’environ 0,1 percent.

Introduction

The measurement of small gas flows is an important technology in semiconductor industries, environmental measurements, gas mixture techniques, and chemical analysis. Higher accuracy flow measurements have been required in these fields. Therefore, reliable and accurate primary standards are indispensable, and many national laboratories have made significant efforts to establish primary standards by their own methods. Intercomparison tests, which compare the standards of one national laboratory to those of another national laboratory, are considered essential to detect and eliminate systematic errors in calibration facilities. Furthermore, intercomparison tests serve to verify uncertainty analyses and to improve the reliability of flow measurements in different countries.

The NRLM Transfer Standard

The National Research Laboratory of Metrology in Japan (NRLM) has developed a transfer standard using sonic venturi nozzles. The nozzles were calibrated by the NRLM primary standard using the static gravimetric method. The NRLM transfer standard is designed to be easily transported to other laboratories for intercomparison tests. Figure 1 is a schematic picture of the NRLM transfer standard. The sonic venturi nozzle can be changed depending on the desired flow. The transfer standard has two sets of pressure and temperature sensors which allow assessment of damage or drift caused by shipping. After the flow rate is measured using one set of sensors, the measurement is repeated using another set without changing the flow conditions. If the two measurements are within tolerance, it indicates that the flow rate is measured correctly. If not, it indicates a change in the sensors most likely caused by transportation or drift. The data from the sensors are sent to a laptop computer using a GPIB interface to calculate the flow parameters. The results are displayed on the screen and are written to files.

The discharge coefficients of the sonic nozzle were determined for various gases using the NRLM primary standard and were fitted as a function of the theoretical Reynolds number. Here, the theoretical Reynolds number is based on the theoretical mass flow calculated from the nozzle.

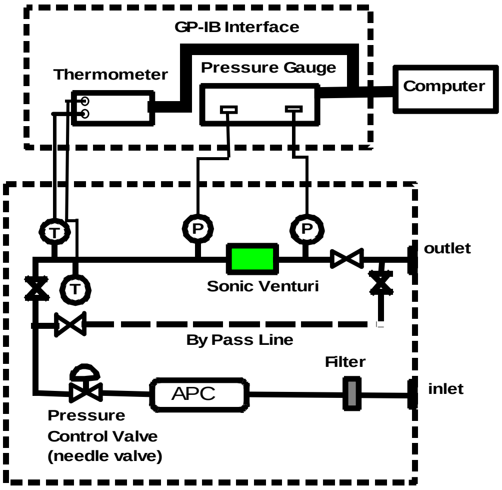

Figure 1. Schematic diagram of the NRLM transfer standard.

The uncertainty of mass flow is composed of the uncertainties of the discharge coefficient and of the calculated theoretical mass flow. The uncertainty of the discharge coefficient consists of the uncertainty of the NRLM primary standard, the repeatability of the measurements, the standard deviation of the discharge coefficient curve fit residuals, and the uncertainty of the theoretical Reynolds number due to the measurement of the pressure and the temperature. The uncertainty of the mass flow measured by the sonic venturi nozzle depends on the nozzle, the gas species and the flow rate. The relative expanded uncertainty (k = 2) of the NRLM transfer standard mass flow measurement was less than 0.15 percent over the range of these tests.

The NIST Primary Standard

The NIST Fluid Flow Group uses piston provers, bell provers and a Pressure-Volume-Temperature-time system to cover flows from 0.04 g/min to 86 000 g/min. The details of operation and uncertainty analyses for the Fluid Flow Group primary standards have been presented previously [2] and will only be briefly described here. In the piston prover system (Figure 2), the metered gas is diverted by valving into a glass cylinder to raise a mercury sealed piston. As the piston rises through the cylinder, it successively starts and stops a timer by blocking the light passing through machined slits at the ends of the collection volume. The temperature and pressure of the gas entering the collection volume are measured with a temperature sensor and an absolute pressure gauge. The temperature and pressure are used to calculate the density of the collected gas, and the density is used to convert the measured volumetric flow rate into a mass flow rate.

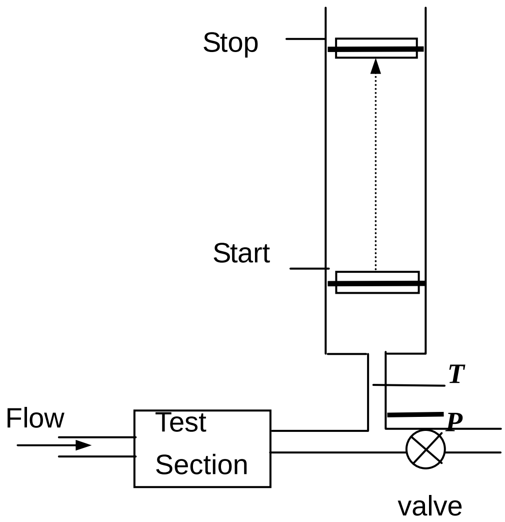

Figure 2. NIST Fluid Flow Group piston provers.

The mass flow measurements made with the piston provers are subject to uncertainties in the determination of the gas density, collection volume, and the collection time. A summary of the uncertainty analyses for the medium and large piston provers used in the April 1996 comparison with NRLM are shown in the following table. The relative standard uncertainty for each of the piston provers was 0.09 %, and the expanded uncertainty (k = 2) was 0.18 %.

Table 1. Summary of uncertainties for the NIST Fluid Flow Group piston provers in April 1996.

Uncertainty Category	Relative Standard Uncertainty	
	Medium (%)	Large (%)
Collection Volume Density	0.061	0.053
Temperature	0.047	0.037
Pressure	0.022	0.022
Fitting Function	0.029	0.029
Experimental Data	0.012	0.012
Collection Volume	0.011	0.033
Cylinder Diameter	0.009	0.032
Collection Length	0.001	0.001
Thermal Expansion	0.006	0.006
Collection Time	0.058	0.061
Timer Calibration	0.001	0.001
Timer Actuation	0.057	0.057
Piston Rocking	0.012	0.023
Storage Effects	0.007	0.001
Combined Relative Standard Uncertainty	0.09	0.09
Expanded Relative Standard Uncertainty (k = 2)	0.18	0.18

BNM-LNE Primary Standard

The BNM-LNE has implemented a gravimetric gas flow standard with real time weighing techniques. Figure 3 shows the principle of the primary gravimetric flow bench. The gas flow is supplied by a gas cylinder under pressure located on the balance. Pressure is controlled by a line pressure regulator and a needle valve. The Molbloc-Molbox system manufactured by DH-Instruments is an integral part of the gravimetric bench. It is used for monitoring the stability of the flow. When the gas flow is regarded as stable, a measurement is initiated. At the start time, the actual mass value of the balance is taken by a PC and the measurement of time interval begins. After the desired mass of gas has been discharged from the cylinder, the computer interrupts the timer and the time interval is determined.

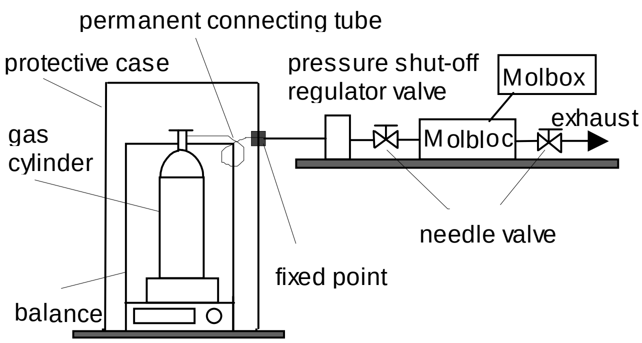

Figure 3. Diagram of the mass flow primary standard of the BNM-LNE.

The Molbloc can be used to check if the mass flow remains stable over the whole time period. This allows the mass flow to be calculated as the quotient of the mass difference and the time interval.

The resolution and the repeatability of the balance and the timer have an important influence on the uncertainty of the measurement. At the BNM-LNE, different electronic balances are used to measure the mass loss of gas with an appropriate resolution. For the measurements of flow between 0.2 mg/sec and 5 mg/sec, a balance of 1.2 kg capacity with a resolution of 10 mg is used. For flows between 5 mg/sec and 0.2 g/sec, a balance of 8 kg capacity with a resolution of 10 mg is used. For higher flows up to 1 g/sec, a balance of 16 kg capacity with a resolution of 0.1 g is used. The timer resolution is 0.1 sec. The relative expanded uncertainty (k = 2) of the mass flow between 0.2 mg/sec and 1 g/sec measured by the system is 0.2 percent.

The OFMET Primary Standard

The OFMET primary volumetric standard (Figure 4) consists of a set of three precision machined glass cylinders. In these cylinders mercury-sealed pistons are raised by inflowing inert and dry gas. The height (Δh) of each piston is measured by means of a miniaturised laser interferometer with a movable spherical reflector in the centre of the piston. With the measurement of the travelling time (Δt) of the piston, the gas temperature (T_g) in each tube, and the gas pressure (sum of ambient absolute pressure (p_a) and the differential pressure (p_b)), the gas volume flow rate at standard conditions (Q_V) is calculated as follows:

$$Q_V = \frac{1}{4} \cdot D^2 \cdot \pi \cdot \frac{\Delta h}{\Delta t} \cdot \frac{\left(p_a + p_b\right) \cdot T_s}{p_s \cdot T_g},$$

with D being the inner diameter of the cylinder. Assuming ideal behaviour of the gases in the very limited temperature and pressure range, the last term in this equation converts the volume flow rate to standard conditions (T_s = 273.15 K; p_s = 101.325 kPa). The mass flow rate (Q_m) is calculated with:

$$Q_m = \rho_s \cdot Q_V$$

where ρ_s is the mass density of the measured gas at standard conditions.

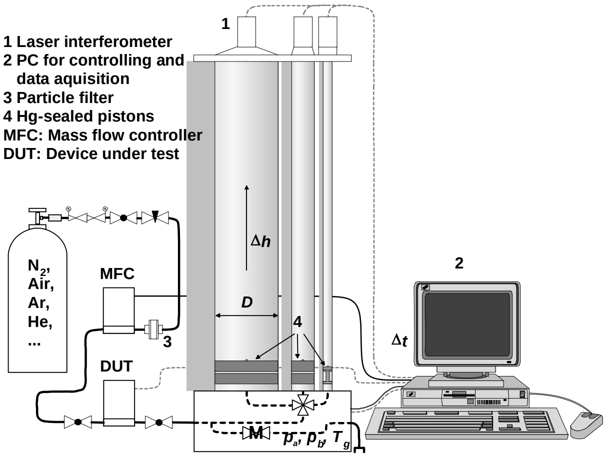

Figure 4. Schematic of the OFMET primary volumetric small gas flow standard

The gas flow covered by the three tubes ranges from 3 cm^3/min to 30 dm^3/min referred to standard temperature and pressure which corresponds to 3.75 mg/min and 37.5 g/min. The relative expanded uncertainty (k = 2) of the measurement results have been evaluated according to international guidelines [6] and are within 0.13 percent for the large and medium tubes which were used for the intercomparison.

The PTB Primary Standard

The mass flow calibration system realised at PTB uses the principle of observing the mass loss of a gas cylinder that is permanently connected with the mass flowmeter (DUT in Figure 5) during the measuring time. Usually a mass flow controller (MFC) is used for controlling the gas flow through the DUT, but other controlling units can also be used. The balance used in the calibration system is a mass comparator with a capacity of 10 kg and a resolution of 1 mg. The time scale has a resolution of 1 ms.

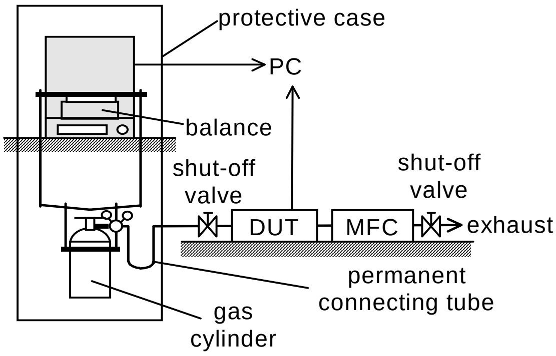

Figure 5. Schematic of the mass flow calibration system of PTB.

The mass values of the balance and the corresponding time values are recorded continuously (every second) over the whole measuring time. The mass flowrate is calculated using regression analysis of the data set composed of the air-buoyancy-corrected mass values and the corresponding time values. If the mass flowrate is constant, the mass of the gas cylinder will decrease linearly with time. The mass flow rate is then given as the slope of the linear regression function.

The advantage of this system is its capability of assessing the stability of the mass flow using the (off-line calculated) residuals of the mass values. If the residuals do not form a uniform band around zero, the flow was unstable. Such instabilities are, for example, caused by variations of the mass flow controlling system. One deficiency of this calibration system is the time measurement (resolution 1 ms) that is still unsatisfactory. This is why it is not possible to measure mass flow rates over 2.5 g/min with satisfactory uncertainties. The continuously measuring dynamic-gravimetric mass flow calibration system of PTB allows mass flow rates between 0.03 g/min and 2.5 g/min to be measured. The relative expanded uncertainties (k = 2) of mass flow rates higher than 0.5 g/min are 0.13 %. For mass flow rates smaller than this amount, the relative expanded uncertainties increase up to 0.6 % for mass flow rates of 0.05 g/min.

Results and Discussion

For the intercomparison tests, the NRLM transfer standard was connected in series to the primary standards of each national laboratory. The transfer standard was connected on the upstream side at NIST and OFMET, and the downstream side at LNE and PTB. The results are summarized in Fig. 6. The horizontal axis is the mass flow measured by the NRLM transfer standard while the vertical axis is the percent difference of the flow measurements of the various national laboratories from the NRLM transfer standard. As previously described, the NRLM transfer standard was calibrated via the NRLM primary standard and there is no significant calibration offset between them.

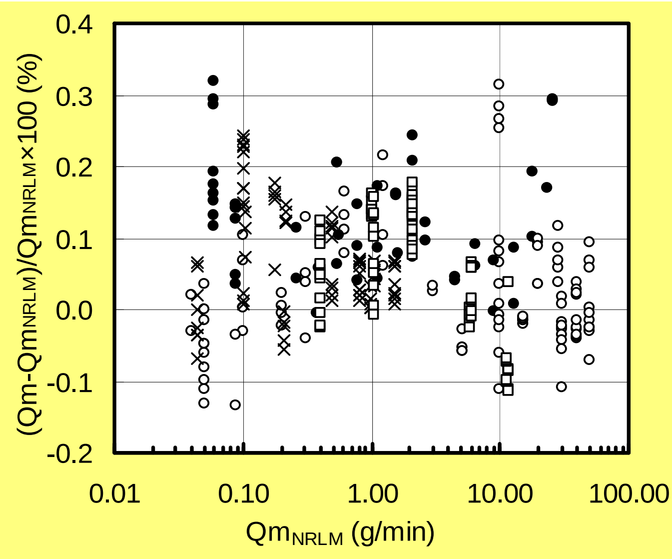

Figure 6. The results of the intercomparison tests. (°:BNM-LNE, □:NIST, •: OFMET, ×:PTB)

In general, the results show good agreement for the whole flow range tested. This agreement is amazing when one considers that the primary standards were established based on a variety of methods by independent researchers. Closer examination of the data shows that the results between 0.2 g/min and 10 g/min agree within 0.2 %. Outside of this flow range, the variation of the results is greater than 0.2 % but smaller than 0.3 %. In the flow range near 1 g/min, the results agree within 0.1 %.

The present intercomparison tests lead to important suggestions for future work in the field of small gas flow measurement; although the efforts up to now have been quite successful, it is necessary to establish flow standards and perform comparisons in the more difficult lower flow range, especially less than 0.1 g/min. The design and construction of much more accurate and reliable standards in this very small flow range will require further cooperation between national laboratories.

References

[1] S. Nakao and J. Barbe, *Intercomparison Tests of BNM-LNE with NRLM for Small Mass Flow Rates of N2*, Ar and He, in the proceedings of FLOMEKO98, 1998, pp. 517-522.

[2] J. Wright and G. Mattingly, *NIST Calibration Services for Gas Flow Meters: Piston Prover and Bell Prover Gas Flow Facilities*, NIST Special Publication 250-49, August 1998.

[3] J. Wright, G. Mattingly, S. Nakao, Y. Yokoi, and M. Takamoto, *International Comparison of a NIST Primary Standard with an NRLM Transfer Standard for Small Mass Flow Rates of Nitrogen Gas*, Metrologia, 35, pp. 211-221, 1998.

[4] J. Barbe and A. Marschal, *Traceability of gas flow measurements*, in the proceedings of Metrology Congress, 1997, France, pp. 517-523

[5] Encyclopedie des gaz, L'AIR LIQUIDE, Elsevier, 1976.

[6] *Guide to the Expression of Uncertainty in Measurement*, ISO Geneva, 1995.

[7] D. Knopf, W. Richter, *Highly Accurate Calibration Gas Mixtures by Dynamic-Gravimetric Preparation*, PTB-Mitteilungen, 108, March 1998, pp. 201-205.