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NIST–IMGC comparison of gas flows below one litre per minute

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Abstract
We compared the IMGC and NIST standards for small gas flows at the IMGC. The IMGC standard is a recently developed primary flow meter that extracts a large piston out of a temperature-controlled chamber. Controlling the piston's speed holds the pressure in the chamber constant. The NIST standard is a recently developed transfer standard that was calibrated against two primary standards at NIST. The first NIST primary extracts a large piston out of an oil-filled chamber in which a metal bellows is suspended. Controlling the piston's speed holds the pressure in the bellows constant. The second NIST primary is a static gravimetry method.

The results include 49 nitrogen flow rates from 0.22 μmol s^{-1} to 770 μmol s^{-1} (1 μmol s^{-1} = 1.3448 cm^3 min^{-1} of an ideal gas at the 'standard' conditions of 0 °C and 101 325 Pa). For all but one of the comparisons, the agreement between IMGC and NIST is better than 0.06%.

1. IMGC flow standard

The IMGC prover (figure 1) is a primary standard that uses a massive, motor-operated piston to change the volume of a temperature-controlled chamber. When the piston is introduced into the chamber, the motor speed is controlled to deliver gas at a constant rate. When the piston is extracted, the motor speed is controlled to hold the chamber at a constant pressure, which is usually near atmospheric. The maximum displacement of 266 mm and the piston's diameter of 120 mm define a maximum stroke of 3 L. A coordinate measuring machine determined the piston's area of cross-section. A commercial laser interferometer measures through a glass window the piston's displacement, or more exactly the true quantity of interest: the displacement of the piston's bottom face referred to the base plate of the measuring chamber. This arrangement avoids all errors caused by thermal expansion of mechanical parts along the axial direction.

Circulation of water through the chamber wall and the piston body controls the chamber's temperature to $\pm$10 mK. Controlling the chamber near room temperature minimizes the chamber's temperature gradients. Two platinum resistance thermometers (not shown in figure 1) measure the average temperature. They were installed inside two vertical grooves (milled into the chamber's inner wall) to allow close contact with the measured gas.

The prover's uncertainty depends on the flow rate and the piston stroke. According to present estimates, using the full stroke lowers the (standard) uncertainty to 0.013% for the prover's full range from below 1 cm^3 min^{-1} to 1000 cm^3 min^{-1}. Using a smaller stroke increases the uncertainty; however, uncertainties lower than 0.02% can be achieved at all flow rates with reasonable test durations (from minutes up to 3 h). The IMGC prover is described in more detail elsewhere [1].

2. NIST flow standard

2.1. Primary standards

The NIST transfer standard was calibrated at NIST against two primary standards. The first NIST primary standard is a constant-pressure flow meter (CPFM), whose largest moving part is a steel piston that moves into or out of an oil-filled chamber. Suspended in the oil chamber is a stainless steel bellows whose interior is connected to the gas flow path. A displacement of the piston into the oil chamber reduces the bellows volume, thereby causing flow out of the bellows. The

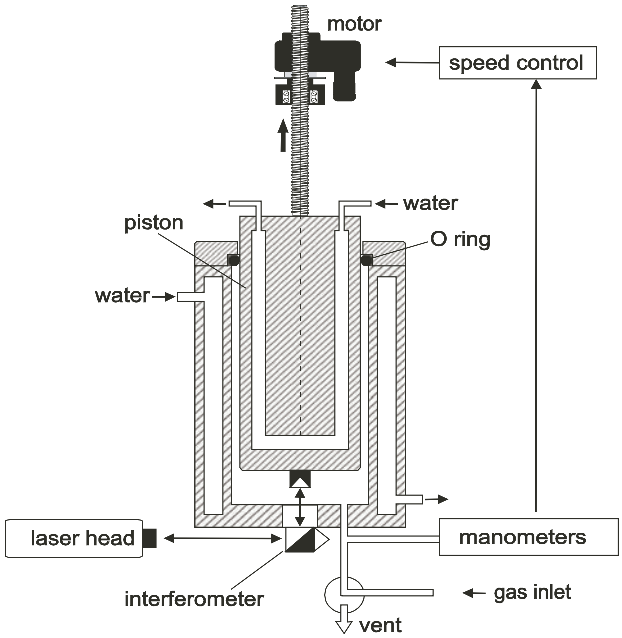

Figure 1. Schematic diagram of the IMGC primary standard. The piston rises at a controlled speed as the gas enters into the measurement chamber; both temperature and pressure are kept constant.

maximum displacement of 110 mm and the piston's diameter of 102 mm define a maximum stroke of 0.9 L. A coordinate measuring machine determined the piston's area of cross-section. A commercial laser interferometer measures the piston's displacement.

During the calibration of the transfer standard, gas flows at an approximately constant rate through the transfer standard and into the CPFM. (A reverse flow out of the CPFM is possible.) The piston speed is controlled to hold the bellows at a constant pressure, which can be as large as 1 MPa, but is usually near atmospheric. The moles of gas accumulated in the CPFM bellows is compared to the integral of the molar flow rate through the transfer standard. The bellows temperature is always near room temperature because it is not controlled. The gas temperature is inferred from the temperature of the aluminium housing, which is measured by several platinum resistance thermometers.

The second NIST primary standard is a gravimetric flow meter (GFM). Gas flows from a small gas bottle through the flow transfer standard. The mass change of the gas bottle is compared to the integral of the mass flow rate through the transfer standard. The weight w_{ref} of a reference bottle is measured as well as the weight w_{gas} of the gas bottle. The mass change is calculated from the change of the difference $w_{gas} - w_{ref}$. The reference bottle's similar mass reduces errors due to balance drift and non-linearity, and its similar volume eliminates the need for buoyancy corrections. The NIST primary standards are described in more detail elsewhere [2].

2.2. Transfer standard

The NIST transfer standard (figure 2) is a laminar flow meter, which means that it uses pressure measurements to

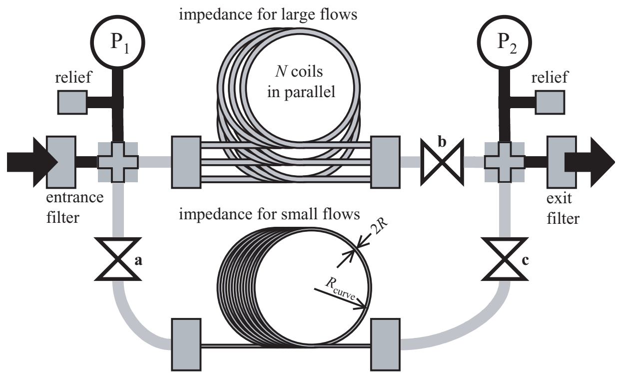

Figure 2. Schematic diagram of the NIST laminar flow transfer standard. Valves a, b, and c direct gas through one of the two installed impedances, and the flow rate is determined from temperature and the pressures P_1 and P_2.

determine the rate of non-turbulent flow through an impedance. Each of the three flow impedances used in this comparison was made from quartz capillary tubing manufactured for gas chromatography. Commercial pressure gauges based on a vibrating quartz flexure measure the input and output pressures P_1 and P_2. The entrance gauge was 'tared' to the exit gauge at zero flow rate, which caused the accuracy of the difference $P_1 - P_2$ to be limited only by the gauge's reproducibility. A temperature-controlled air bath housed the flow impedance, the pressure gauges and two platinum resistance thermometers, all of which were attached to an internal frame.

The transfer standard allowed a comparison between the CPFM and GFM at flow rates from $0.08\,\mu\text{mol s}^{-1}$ to $120\,\mu\text{mol s}^{-1}$ (0.1 to 160 standard $\text{cm}^3\,\text{min}^{-1}$). The typical difference between the two primaries (0.06%) exceeded their estimated separate uncertainties of $\leqslant 0.03\%$. Therefore the true value of flow rate was assumed to lie somewhere in between with a rectangular probability distribution. The resulting uncertainty dominated the standard uncertainty of the transfer standard, which was approximately 0.04% at all flow rates. The NIST transfer standard is described in more detail elsewhere [3].

3. Procedure

3.1. Transportation of the transfer standard

The comparison was conducted at the IMGC during five days in 2001. Commercial plane and truck carriers delivered the NIST transfer standard package from Gaithersburg to Torino without damage. The 50 kg package included a laptop computer, necessary electronics, and miscellaneous supplies as well as the transfer standard. Only one of the three impedances was installed during shipping; the other two were hand carried.

3.2. Measurement preparation

Figure 3 shows the relative arrangement of the two flow standards. Nitrogen gas of 99.9995% purity was directed through a pressure regulator and the NIST flow standard to

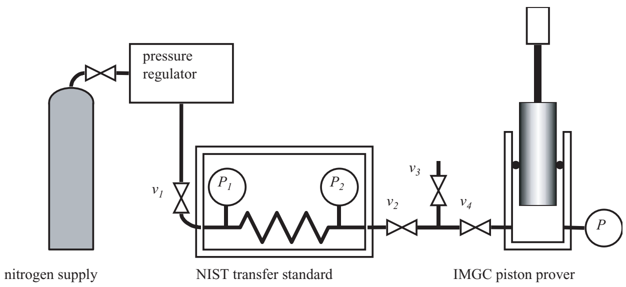

Figure 3. Schematic diagram of the flow comparison.

the IMGC flow standard. For the smallest flow rates, a second, more precise, pressure regulator was added. The NIST standard was the upstream standard because it required pressure drops as large as 200 kPa for its larger flow rates. Thus, the IMGC prover always operated in its constant-pressure mode.

The comparison was conducted in a room whose temperature-controlled walls usually stabilize the air temperature to within 0.3 K. Reducing the connecting volume between the two flow standards to 2 cm^3 further reduced errors due to changes of room temperature.

Multiple cycles of pressurization and flushing removed air from the system. The lack of a significant pressure change upon pressurizing the transfer standard and closing valves v_1, v_3 and v_4 verified the absence of leaks. During the comparisons, the transfer standard's pressure transducers were tared against each other at least once daily; the uncorrected value of $P_1 - P_2$ ranged from 24 Pa to 28 Pa. The IMGC pressure gauge reading P was compared with the NIST average $(P_1 + P_2)/2$ on two separate days; both comparisons agreed within 4 Pa.

3.3. Measurements

For each comparison, the pressure regulator was adjusted to give the desired flow rate with the NIST standard exhausting to atmosphere. Closing valve v_3 (figure 3) directed the flow into the piston prover; its constant-pressure operation caused the piston to rise. After damped oscillations, the piston's velocity reached its asymptotic value within typically 20 s to 60 s. The piston speed was considered to have achieved its final and constant value when fluctuations of the absolute pressure were less than ±1 Pa. The stability of gas temperature in the measuring chamber was assessed *a posteriori*: absolute values of temperature drifts during each test were normally lower than 10 mK (the average being 5 mK).

After achievement of steady state conditions, the piston prover's displacement, as well as gas pressure and temperature, were recorded at the initial and final measurement times. The transfer standard's pressures and temperatures were recorded during the same interval. The full piston stroke was used for only 15 of the tests at larger flow rates. Smaller strokes were used to reduce the duration of the tests at smaller flow rates. (The smallest flow rate caused a piston speed of only 17 mm per day.) Each of the flow impedances was used on at least two different days. Table 1 shows the order of the flow comparisons.

Table 1. The sequence of flow comparisons. The column indicates the flow impedance used in the transfer standard for the groups of comparisons. Each entry denotes the number of comparisons in that group.

	Flow rates		
Date	Small	Medium	Large
27 Sept.		14	
28 Sept.		1	
			17
		3	
	2		
29 Sept.	4		
30 Sept.	3		
1 Oct.		8	
			5
Total	9	26	22

4. Analysis

4.1. IMGC piston prover

The molar flow rate measured by the IMGC piston prover is

$$\dot{n}_{\mathrm{IMGC}} = \frac{\rho_2(V_{\mathrm{d}} + A\Delta x) - \rho_1 V_{\mathrm{d}}}{Mt} \qquad (1)$$

where ρ_1 and ρ_2 are the initial and final gas densities, V_{d} is the dead (or initial) volume, A is the piston's area of cross-section, and Δx is the piston's displacement during time t. The gas density is given by

$$\rho = \frac{MP}{ZR_{\mathrm{gas}}T} \qquad (2)$$

where P, M, T, and Z are the gas's pressure, molar mass, temperature, and compressibility factor, and R_{gas} is the universal gas constant.

4.2. NIST transfer standard

The molar flow rate measured by the NIST transfer standard is

$$\dot{n}_{\mathrm{NIST}} = \frac{\pi(P_1^2 - P_2^2)R^4}{16R_{\mathrm{gas}}T\eta L}[1 + \text{corrections}]f_{\mathrm{C}}(\mathrm{De}) \qquad (3)$$

where P_1 and P_2 are the entrance and exit pressures, R and L are the capillary radius and length, and η is the gas viscosity. Each of the five 'corrections' in the brackets is less than 0.2%. They account for the gas's non-ideality, slip at the capillary walls, the pressure drop at the capillary entrance, the effect of gas expansion on the velocity distribution, and thermal effects within the capillary. The function $f_{\mathrm{C}}(\mathrm{De})$ accounts for the centrifugal effects due to the coiled capillary. Further details are available in [3].

For each impedance, the model represented by (3) had only one free parameter, the capillary radius R. The calibration by the primary standards determined R. (For the medium impedance, an independent method found the same value of R within the method's resolution of 0.1%.) Flow measurements made at NIST after the comparison verified that the value of R for each impedance had not changed.

Two adjustments to the model were made after the comparison was completed. This included the fifth correction

Table 2. Uncertainty parameters for the IMGC piston prover (see footnote 3).

Quantity	Symbol	Value	Uncertainty	
Piston area	A	11 298 mm^2	u_A	1.1 mm^2
Piston displacement	Δx	14 mm to 200 mm	$u_{\Delta x}$	1 μm
Temperature	T	≅296 K	u_T	20 mK
Pressure	P	98 900 Pa to 99 320 Pa	u_P	5 Pa
Time	t	86 s to 52 000 s	u_t	5 ms
Dynamic leaks, gasket distortion	—	—	u_{leak}	10 mm^3
Initial volume	V_1	360 cm^3 to 1440 cm^3	—	—
Final volume	V_2	514 cm^3 to 3120 cm^3	—	—
Measured volume	$V_2 - V_1$	75 cm^3 to 2400 cm^3	See u_A and $u_{\Delta x}$	

to (3) and an adjustment to R required by improvements to the models of the NIST primaries. Because the net changes were small (i.e. $\leqslant$0.03%), the original flow results were adjusted by an appropriate linear function of flow rate.

4.3. Uncertainty

The relative standard uncertainty of the IMGC flow standard estimated at the time of the comparison was[3]:

$$\left(\frac{u_{\dot{n}}}{\dot{n}}\right)_{\text{IMGC}} = \left[\left(\frac{u_A}{A}\right)^2 + \left(\frac{u_{\Delta x}}{\Delta x}\right)^2 + \frac{u_{\text{leak}}^2 + [(u_T/T)^2 + (u_P/P)^2](V_1^2 + V_2^2)}{(V_2 - V_1)^2} + \left(\frac{u_t}{t}\right)^2\right]^{1/2} \quad (4)$$

where u_{leak} is the estimated upper bound of the dynamic leak, and V_1 and V_2 are the initial and final volumes. Here, the uncertainty of temperature T, for example, is indicated by u_T. Table 2 gives the values of the quantities and the uncertainty parameters for (4).

The denominator of the third term of (4) makes clear that a small uncertainty requires a large metred volume ($V_2 - V_1$). Achieving state-of-the-art accuracy requires at least 200 cm^3. Similarly, the numerator shows that the initial volume must be kept as small as possible. The initial volume V_1 is made of three components: (a) the internal volume of the chamber, manometers and associated plumbing up to the input and exhaust valves v_2 and v_3 (≅357 cm^3, see figure 3); (b) the almost negligible volume of fittings connecting the two standards (≅2 cm^3, not shown in figure 3); (c) the variable volume swept by the piston during its acceleration and stabilization phase.

The relative uncertainty of the NIST flow standard is

$$\left(\frac{u_{\dot{n}}}{\dot{n}}\right)_{\text{NIST}} = \left[4\left(\frac{u_R}{R}\right)^2 + 4\left(\frac{u_P}{P_1 + P_2}\right)^2 + 2\left(\frac{\delta P}{P_1 - P_2}\right)^2 + \left(\Delta_\eta \text{De}^4 \frac{u_\eta}{\eta}\right)^2 + \Delta_T^2\right]^{1/2} \quad (5)$$

[3] A later, improved analysis [1] took into account the correlation existing between initial and final measurements of temperature and pressure in the IMGC prover, as well as minor changes in early evaluations of some uncertainty components. Using it would have decreased the combined uncertainty by less than 12%.

Table 3. Average contributions of the two standards to the combined standard uncertainty (see footnote 3).

	Flow rates		
	Small	Medium	Large
NIST	0.040×10^{-2}	0.040×10^{-2}	0.040×10^{-2}
IMGC	0.028×10^{-2}	0.032×10^{-2}	0.019×10^{-2}
Combined	0.048×10^{-2}	0.051×10^{-2}	0.045×10^{-2}

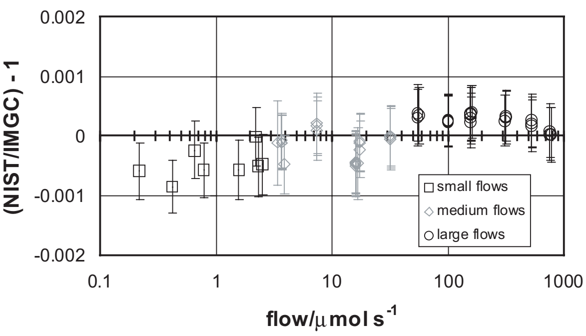

Figure 4. Comparison results: the three flow ranges correspond to the three flow impedances used in the NIST transfer standard. The error bars represent the combined standard uncertainty given by (6).

where δP is the pressure gauge's reproducibility, Δ_η is the sensitivity of the centrifugal correction to the viscosity uncertainty u_η, and Δ_T is the contribution of temperature gradients within the transfer standard. The dominance of the capillary radius uncertainty u_R caused the flow uncertainty to be approximately 0.04% for all flows in this comparison (see [3] for more details).

The combined standard uncertainty of the two interfaced standards (and hence of the comparison between their results) is:

$$\left(\frac{u_{\dot{n}}}{\dot{n}}\right)_{\text{combined}} = \left[\left(\frac{u_{\dot{n}}}{\dot{n}}\right)^2_{\text{IMGC}} + \left(\frac{u_{\dot{n}}}{\dot{n}}\right)^2_{\text{NIST}}\right]^{1/2}. \quad (6)$$

Table 3 shows the average contributions of the IMGC (primary) and NIST (secondary) standards to the combined uncertainty. For the IMGC standard, the medium and small flows had larger average uncertainties because they used shorter piston strokes to reduce the duration of the tests. The uncertainty for medium flows was larger than for small flows because the stabilization portion of the stroke for medium flows was larger, which gave a less favourable ratio between the metered volumes and initial volumes.

5. Results

Eight of the 57 comparisons listed in table 1 were excluded from the results. One of the small flow comparisons differed by 0.1% from the others. It was excluded because the difference was apparently caused by an unusual 0.4 K shift of room temperature that occurred during the overnight run. Six of the medium flow comparisons were excluded because we had used either a deliberately small metered volume (<150 cm^3) or an unnecessarily large dead volume. Such conditions, which caused a large uncertainty, were used to find any unexpected dependence on metered volume or on dead volume; none was found. Finally, the first large flow comparison was excluded due to a lack of temperature equilibrium.

Figure 4 shows the final comparison results in the form $\dot{n}_{\mathrm{NIST}}/\dot{n}_{\mathrm{IMGC}} - 1$. The 49 nitrogen flow rates range from 0.22 μmol s^{-1} to 770 μmol s^{-1}. For all but the next-to-smallest flow rate, the agreement between IMGC and NIST was better than 0.06%. Another way to express this agreement is to divide the difference between laboratories by its expanded combined uncertainty, $U_{\mathrm{combined}} = 2u_{\mathrm{combined}}$:

$$E = \frac{\dot{n}_{\mathrm{NIST}} - \dot{n}_{\mathrm{IMGC}}}{2u_{\mathrm{combined}}}. \qquad (7)$$

Obtaining $|E| < 1$ suggests that u_{combined} was estimated correctly. For the small, medium, and large flows, the respective average values of E are -0.5, -0.2, and +0.3. Further details and tables of numerical data are available from the authors.

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