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Bilateral comparison of gas microflow primary standards in the range between 0.12 mg/min and 2.4 mg/min

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ABSTRACT

A bilateral comparison of small gas flow rates in the range between 0.12 mg/min and 2.4 mg/min was performed between INRiM (Italy) and LMPS (Slovenia). The primary standard at INRiM is a type of a motor driven piston prover. The primary standard at LMPS is a pVTt method with a static mass determination and flying start-stop. The transfer standard main part are two capillary leaks, with measuring ranges up to 0.25 mg/min and 2.5 mg/min respectively. A Hagen-Poiseuille equation with a constant geometric factor is used to calculate the mass flow rate. The comparison was carried out at five measurement points with the large capillary leak and at two points with the small capillary leak. The results show that there are statistically significant differences between the measurements of the two laboratories. The estimated uncertainties are also too large to confirm the target CMCs, as the repeatability of the measurements was too weak.

1. Introduction

Lately a strong tendency of the industry and government to reduce the leakage of the products has developed in order to boost efficiency and reduce ecological footprint of the mankind. Based on that, metrological capabilities of research institutions in the field of small flow rate measurement are gaining relevance. An additional driver of infrastructure development is the planned transition to hydrogen of the European Union energy sector. The metrological infrastructure for this transition is still in development [1]. The laboratory LMPS recently developed a primary standard for gas micro flow with a measuring range from 0.12 mg/min to 12 mg/min. In scope of the EURAMET project Met4H2 the standard is being prepared to measure hydrogen flow rates. In this context, a preliminary comparison based on nitrogen flow measurement was performed with INRiM.

The comparison was realized in the flow rate range between 0.12 mg/min and 2.4 mg/min. LMPS used the pVTt primary standard, which can be characterized by relatively short measuring times [2,3], while INRiM uses the motor driven piston prover [4,5]. For the comparison the transfer standard based on two fixed geometry capillary leaks was developed. The flow rate through the capillary leak depends on the pressure drop across its capillary element. The comparison was performed with nitrogen purity above 99.999 %.

2. INRiM primary standard

The INRiM gas flow primary standard is of volumetric type with a changing volume. It uses a motor driven piston to change the volume of the thermally controlled chamber. A piston with a diameter of 120 mm has a maximum travel length of 266 mm and a total displacement of 3 L, with dimensions determined using the coordinate measuring machine. The inset of the piston into the chamber is measured with the laser interferometer through the window below the piston. The position of the piston is motor controlled in a way that keeps the constant differential pressure inside the measurement volume compared to surrounding atmosphere. It can function as a collector (differential pressure kept at zero) of the gas flow or as the gas flow generator (differential pressure up to 1 kPa). Using cooling channels within the piston and the chamber wall the entire system is thermally stabilized. The uncertainty of the measurement depends on the piston stroke, with smaller stroke meaning higher uncertainty. Using measurements up to 3 h the achievable expanded uncertainty below 0.05 % is predicted for flow rates between 0.1 ml/min and 2 l/min [4,5].

The transfer standard was connected in series with the INRiM reference standard that was operating as a collector of the gas flow, keeping a stable zero differential pressure. The readings from the transfer standard were recorded and averaged for the whole measurement time of the INRiM standard. The setup is presented in Fig. 1.

3. LMPS primary standard

The LMPS gas flow primary standard is of volumetric type with a constant volume, usually abbreviated as pVTt (pressure, volume, temperature and time). It has the measurement volume of 102 ml, from which most of the volume represents a calibrated cylinder of 32 mm diameter and 117 mm length. In the cylinder wall, a platinum resistance probe is inserted to indirectly measure the gas's temperature. A pneumatic diverter diverts the gas flow into the measurement volume where the gas is collected up to maximum differential pressure of 2.5 kPa compared to stable atmospheric reference pressure. The mass flow rate is determined based on the measurement volume size, the density change, determined according to gas pressure and temperature in the measurement volume prior and after the mass collection, and the collection time, determined according to the diverter operation. By

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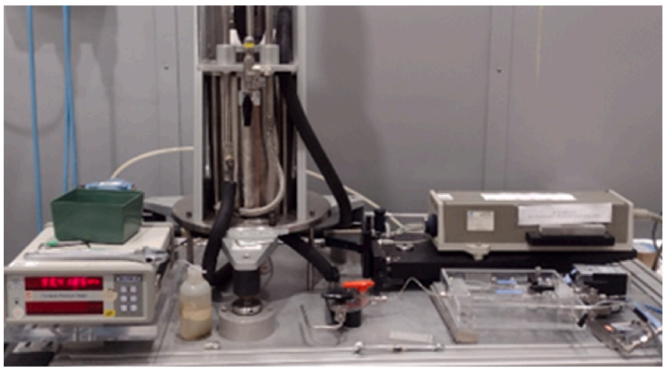


Fig. 1. Measuring system setup at INRiM.

extending the collection time up to 25 min with maximum allowable differential pressure, the achievable expanded uncertainty below 0.2 % of the measured value is predicted for flow rates between 0.12 mg/min and 12 mg/min [2,3].

The transfer standard was connected in parallel with the LMPS reference standard, with constant mass flow rate alternatively directed to the reference standard or to the transfer standard. The readings from the transfer standard were recorded before the measurement with the reference standard. The setup is presented in Fig. 2.

4. Transfer standard and ILC procedure

The transfer standard consists of a small and a large capillary leak, with measuring ranges up to 0.25 mg/min and 2.5 mg/min, respectively, a differential pressure transducer measuring pressure drop across the capillary leak p_{dif}, a temperature probe measuring gas inlet temperature T and a control unit for data acquisition and PC communication, with an additional absolute pressure sensor measuring ambient pressure p_{amb} (Fig. 3). Upstream of each capillary leak, a shut-off valve is placed to control which capillary leak the flow rate is directed through. The flow in both capillary leaks is treated as laminar, so a Hagen-Poiseuille law with the constant geometric factor K is used to calculate the volume flow rate. With the density defined at average pressure in the capillary leak, we obtain the mass flow rate as:

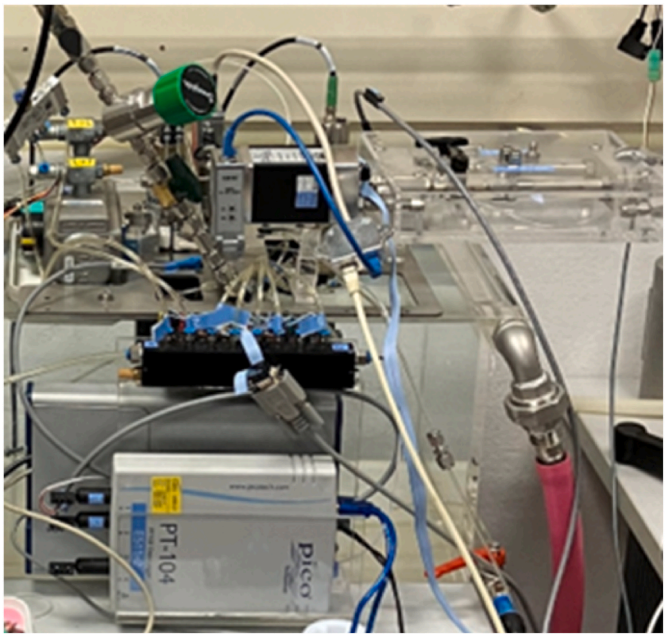


Fig. 2. Measuring system setup at LMPS.

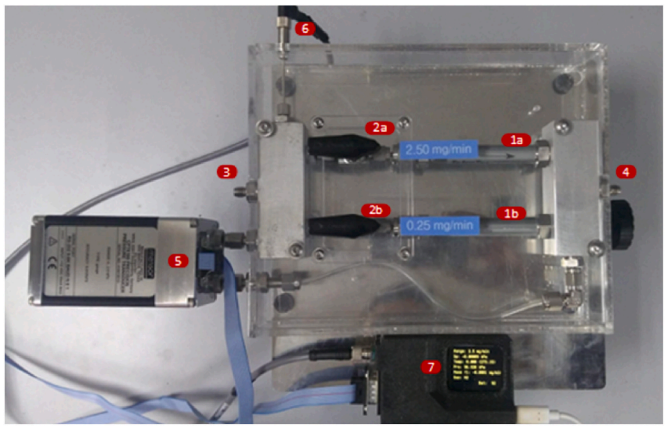


Fig. 3. Photo of a transfer standard. Labelled items: 1 – capillary leaks, 2 – shut off valves, 3 – flow inlet, 4 – flow outlet, 5 – differential pressure transducer, 6 – temperature probe, 6 – control unit with absolute pressure sensor.

$$q_m = K \frac{p_{dif}\left(p_{dif} + 2\,p_{amb}\right)}{R_{spc}\,T\,\mu\,Z}, \tag{1}$$

with R_{spc} is the specific gas constant, $Z = Z\,(T, \bar{p})$ is the compressibility factor and $\mu = \mu\,(T, \bar{p})$ is the dynamic viscosity.

Before the interlaboratory comparison was carried out the experimental analysis of the transfer standard was conducted by the LMPS to define the geometric constants and to evaluate the ambient temperature and pressure effects. No significant effects related to the ambient pressure and temperature changes were observed.

The control unit has a built-in display to show the measured values. Additionally, it sends a message every 2 s via USB connection to the PC that contains all the measured values and the calculated mass flow rate. The control unit was supplemented with two buttons for selection of the fluid and the engaged capillary leak, respectively. The compressibility factor and dynamic viscosity of the gas are calculated on the control unit using linear interpolation. Each of the laboratory reported the following set of values from the transfer standard: the differential pressure p_{dif}^{T} (kPa), the temperature T^{T} (K), the absolute pressure p_{amb}^{T} (kPa) and the mass flow rate q_m^T(mg/min) and from the reference standard: the reference mass flow rate q_m^R (mg/min), the expanded uncertainty of the flow rate $U(q_m^R)$ (k = 2, mg/min), the gas temperature T^R (K) and the gas absolute pressure p_a^R (kPa).

Measurements were conducted from the largest to the smallest flow rate. The flow rate was adjusted to obtain the differential pressure p_{dif} within ±2 % of the nominal values given in Tables 1 and 2. At each measurement point three repetitions were performed. To conduct a single measurement, the gas flow was diverted through the capillary leak and after a stabilization time of 120 s and 500 s for the large and the small capillary leak, respectively, the measured values were recorded. An average of at least 30 s (15 readings) was used as the measured value.

The measurements were first performed by LMPS. Then the transfer standard was transferred to the INRiM to perform their measurements.

Table 1
Measurement points for the large capillary leak with the range up to 2.5 mg/min.

#	Differential pressure [Pa]	App. flow rate [mg/min (sccm)[a]]
1.1	2400	2.4 (1.92)
1.2	1800	1.8 (1.44)
1.3	1200	1.2 (0.96)
1.4	600	0.6 (0.48)
1.5	240	0.24 (0.192)

[a] Standard volume flow rate in cm^3min at 101.325 kPa and 0 °C.

Table 2
Measurement points for the small capillary leak with the range up to 0.25 mg/min.

#	Differential pressure [Pa]	App. flow rate [mg/min (sccm)[a]]
2.1	2400	0.24 (0.192)
2.2	1200	0.12 (0.096)

[a] Standard volume flow rate in cm^3/min at 101.325 kPa and 0 °C.

At the end, the transfer standard was returned to the LMPS to perform an additional set of measurements, necessary for the estimation of the time-drift d of the transfer standard during the comparison. All the measurements were conducted between October and December 2023.

5. Evaluation procedureprocedure

The results of both laboratories, their reported measurement errors e and expanded uncertainties $U(e)$, are evaluated in terms of the normalized error value E_n for each measuring point according to ISO 17043 [6]:

$$E_n = \frac{\bar{e}_{LMPS} - \bar{e}_{INRiM}}{\sqrt{U(\bar{e}_{LMPS})^2 + U(\bar{e}_{INRiM})^2 + \left(\frac{2d}{\sqrt{3}}\right)^2}}, \quad (2)$$

which also includes the effect of time drift d of the transfer standard during the comparison. In order to access the time drift of the transfer standard LMPS repeated the measurements at the end of the comparison. The time drift was calculated as the difference of the corresponding mean values at each measuring point.

The average measurement error $\bar{e}$ for each laboratory is defined as the arithmetic average value of the three repetitions at each measuring point:

$$\bar{e} = \frac{e_1 + e_2 + e_3}{3}. \quad (3)$$

As the reported uncertainties do not include the effects of repeatability, we calculate the expanded uncertainty $U(\bar{e})$ for each measurement point combining the maximum reported expanded uncertainty $U(q_m^R)_{max}$ and the experimental standard deviation of the errors $s(e)$. First, we calculate the standard uncertainty $u(\bar{e})$ in accordance with JCGM 100 [7]:

$$u(\bar{e}) = \sqrt{\left(U(q_m^R)_{max}/2\right)^2 + s(e)^2}. \quad (4)$$

Expanded uncertainty is then calculated by multiplying the standard uncertainty by the coverage factor k:

$$U(\bar{e}) = k\, u(\bar{e}). \quad (5)$$

The coverage factor k is determined by considering the Student's t distribution and the confidence level of 95.45 %:

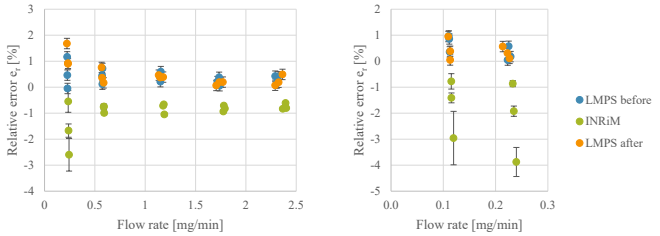
$$k = t_{95.45\%}(\nu_{eff}), \quad (6)$$

where ν_{eff} is the effective degree of freedom, for three repetitions defined as:

$$\nu_{eff} = 2\frac{u(\bar{e})^4}{s(e)^4}. \quad (7)$$

6. Results

The reported relative flow rate errors e_r (relative difference between the transfer standard and the primary standard) of both laboratories are presented in Fig. 4. Reported expanded uncertainty of LMPS is 0.2 % since the reached differential pressure and collection time were always the largest possible. Reported expanded uncertainties of INRiM for flow rates above 0.5 mg/min are between 0.05 % and 0.1 %, while for flow rates below 0.5 mg/min expanded uncertainty increases to 1 %. The collection time of the piston prover was adapted to laboratory external factors, resulting in a larger reported uncertainty at some measurement points. At these points, errors are also substantially larger. The coverage factors of three repetitions were mostly above 3, only for the largest three flow rates of LMPS results the coverage factor was lower, but still above 2.



(a) Values for the large capillary leak. (b) Values for the small capillary leak.

Fig. 4. Relative flow errors including expanded measurement uncertainties $U(q_m^R)$ reported by both laboratories.

The two LMPS measuring sets, performed before and after INRiM measurements, exhibit very small drift. This drift does not exceed 0.0016 mg/min. On the other hand, the scatter of calibration results is substantially larger, especially at lower flow rates.

On the basis of reported relative average errors, relative expanded uncertainties and normalized errors were calculated. The values are presented in Fig. 5. For flow rates below 0.24 mg/min the normalized error values are $|E_n| < 1$, indicating that a statistically significant difference between the measurements in both laboratories does not exist due to the significant scatter of measurement errors. However, for the smallest flow rate of the large capillary leak the higher scatter can be explained by the fact that the differential pressure measured by the pressure transducer of the transfer standard is only about 10 % of its full-

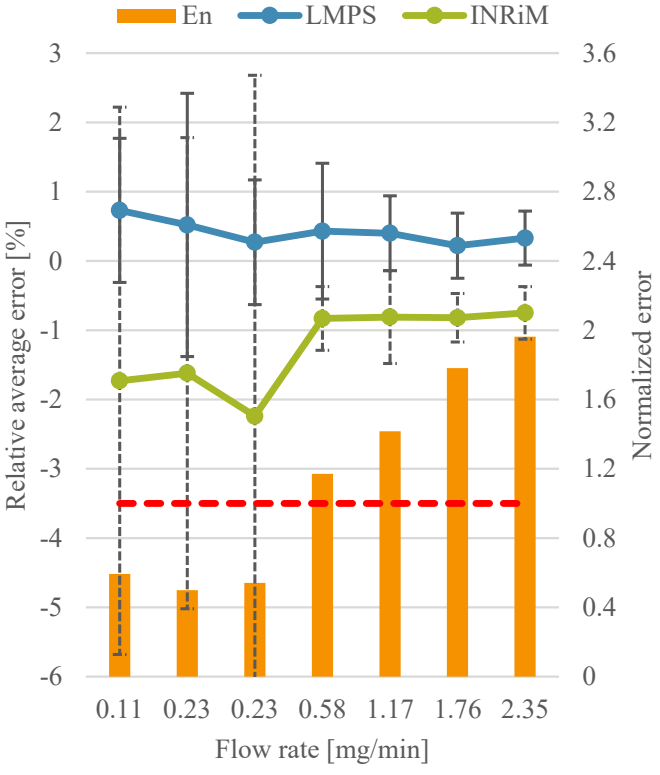


Fig. 5. Relative average errors, relative expanded uncertainties and normalized errors of the comparison.

scale value. For the small capillary leak the pressure response is slow and compensating pressure changes in the ambient atmosphere introduces significant instability of the differential pressure. This arises from the fact that at that flow rate level we were unable to determine if some of the mass flow is accumulating in the volume upstream of the transfer standard. Nevertheless, the observed errors still show a significant amount of discrepancy between the laboratories.

For flow rates above 0.5 mg/min the normalized error values are $|E_n| > 1$, even though the expanded uncertainties $U(\bar{e})$ were about 0.5 % of measured values due to the scatter of the calibration results. There is an almost stable relative error of 1 % between laboratories, which suggests a significant systematic error between the results reported by the laboratories or an unexpected and unaccounted measurement effect on the transfer standard.

7. Conclusions

A comparison in the range between 0.12 mg/min and 2.4 mg/min of nitrogen flow was performed between LMPS and INRiM. Analysis was conducted and normalized errors were calculated according to ISO 17043 [6].

Resulting normalized error E_n was lower than 1 for flow rates below 0.5 mg/min and higher than 1 for bigger flow rates. However, even in former case, the scatter of the measurements results was too high to confirm the target measurement and calibration capabilities of participating laboratories as the assessed coverage factors k for most of the measurements were higher than three.

The resulting discrepancy could be a result of different ambient conditions at both laboratories as micro flow rates are sensitive to the absolute pressure and the temperature conditions that differed notably between the laboratories ($\Delta T = 4$ °C, $\Delta p_a = 1$ kPa). But more likely the discrepancies indicate a systematic error that is not related to the flow rate. Source of a systematic error could occur at one or both laboratories due to several reasons, such as potentially incorrectly evaluated volume of the pVTt method, bad zeroing of the piston position at piston prover, non-optimal temperature measurement position in the pVTt system, unsatisfactory correction for the piston prover dead volume mass accumulation, etc. Both laboratories will have to assess their standards and methods and try to find a possible source of the error.

To reduce the excessive scatter of the calibration results (of both laboratories), the transfer standard should be in the future upgraded with an instream absolute pressure sensor on either side of the capillary leak. Ideally, this absolute pressure should be controlled, as the fluctuations in ambient pressure affect the repeatability of the measurement, especially at the lowest flow rates. This would also provide an opportunity to experimentally evaluate the transfer standard in a broader range of ambient pressure and temperature matching those in both laboratories.

To conclude, to compare and confirm the CMCs of both laboratories in the desired gas flow range, firstly, we must enhance the transfer standard and secondly, we must critically evaluate all possible effect of the primary standards and methods used in comparison, which could lead to the observed systematic errors.

Acknowledgments

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