EXPERIENCE WITH GRAVIMETRIC FLOW STANDARD (GFS) IN VACUUM AND HERMETIC MODE

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Abstract − The Czech national standard of low gas flow is based on a gravimetric flow standard (GFS) principle. It means it defines the flow as the mass loss of gas from a pressure bottle which is continuously weighed. This weighing is performed in an ambient atmosphere which causes that the main source of uncertainty results from the buoyancy correction. This buoyancy correction is complicated by the instabilities of the atmospheric environment and by the fact that volume of the weighed pressure bottle is dependent both on temperature and internal pressure. By modifying the experimental set-up of the GFS utilizing weighing in vacuum, it is possible to suppress the problematic buoyancy effects and moreover to eliminate other impractical side effects such as water vapour condensation on the pressure bottle due to cooling below dew point for the higher flow rates. This presentation describes the practical problems of the vacuum modification of the GFS together with the uncertainty analysis of the modified experimental set-up and the measurement results. The hermetic mode and its main applicability is also introduced.

Keywords: equivalence of national standards, flow metrology, gravimetric flow standard

1. INTRODUCTION

Primary mass low flow standard consisting of dynamic gravimetric system was finished in 2006 from the components supplied and, after that its metrological specification and validation within the EURAMET 806 key comparison was accomplished. The standard itself encompasses pressurized vessel which rests on the precise electronic balance with an automated system for zeroing the balance, calibrating the balance and filling the pressurized cylinder, see Fig. 1 and 2. This system is placed upon a special plate for vibration damping. It is closed in a draft enclosure preventing the effect of air circulation on the pressurized cylinder. It is equipped with a system monitoring the ambient temperature, humidity and pressure conditions. Two pressure regulators are attached to the pressurized cylinder, which ensure a constant pressure in the capillary, interconnecting the pressurized cylinder with a calibrated device. Through this capillary, the measured gas flow passes into the temperature stabilization volume and into the molbloc–molbox system furnished with a control

mass flow meter intended to ensure the flow stability. The GFS – primary low mass flow standard based on gravimetric principle is joined into one total system by a control program which controls individual elements of the standard and calculates the resultant flow with all corrections.
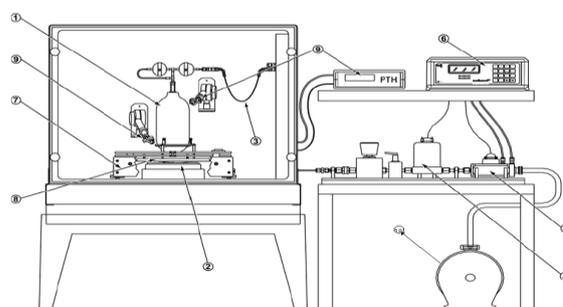

Fig. 1: Schematic of primary low mass flow standard based on the dynamic gravimetric principle - 1 – pressurized cylinder with gas, 2 – electronic balance, 3 – catenary-type flexible manifold, 4 – regulation element, 5 – molbloc, 6 – molbox, 7 – system of automatic filling and tarring of pressurized cylinder, 8 – reference weight, 9 – balance ambient measurement conditions, 10 – air pump.

Fig. 2: Czech primary gravimetric standard of low mass flow (GFS) in atmospheric chamber

The primary standard measures and calculates the mass flow from the decrease of mass of a pressurized cylinder filled with a pressurized gas exhausted through the stabilization

flow meter, which rests on the electronic balance with automated system AMH (automated mass handler) for zeroing the balance, calibrating the balance and placing the pressurized cylinder with compressed gas on the balance during measurement. During the measurement itself, the filled pressurized cylinder is placed on the balance and connected by means of a flexible capillary to the temperature and flow stabilizing system. The constant flow is set with the use of mass flow meter controlled by the control program. After flow stabilization and checking the balance zero, this software starts to measure the decrease of gas mass from the cylinder with a weight indication, from which it directly calculates the mass flow in mg/s, or any other units required. The calculated flow is corrected for the effects upon the system during measurement. It refers especially to the change in buoyancy which affects the cylinder.

There is a permanent demand for decreasing the uncertainty of low gas flow standards from the metrology community. The GFS gravimetric standard accuracy depends on (in)accuracy of buoyancy corrections which are the main contribution to the uncertainty. Placing the pressurized cylinder with gas and the weighing system into a vacuum recipient (see Fig. 3) decreases these influences to practically zero, and results in up to sixfold decrease of uncertainty of the national standard.

Fig. 3: Primary gravimetric standard of low mass flow (GFS) in vacuum chamber

However, many technical problems related to weighing in vacuum had to be solved. Even, weighing in atmosphere, hermetically closed, where at constant air density and omitting the effects of buoyancy on the resultant uncertainty are also rapidly eliminated, would be a significant contribution to the decrease of uncertainty. This would also extend the range of the assembly downwards for calibration of atmospheric freon leaks. The first, a vacuum-type, causes also an automatic increase of the upper range of GFS due to the lack of water vapour, the condensation of which at the weighted pressurized cylinder during a quick cooling of the surface under higher flow, which does not occur in vacuum.

The second, hermetic-type, provides at the same time for a substantial decrease of uncertainties of calibration of atmospheric freon leaks for high ranges, which are also calibrated by means of this device. It would be also possible to use this assembly for weighing in vacuum and for more precise determination of densities of pressure meters of primary standard pressure gauges and weights for piston gauges.

2. APPLICABILITY OF GFS COMPONENTS IN VACUUM

The first stage of solution was to find whether individual components of dynamic gravimeter withstand a vacuum. The main examined parts:

- Electronic balance, this evaluation was critical, since it is the most expensive component of the system. That is why the manufacturer - Mettler was asked, whether this type of balance can be used in vacuum. The manufacturer answered that the balance can withstand the vacuum, but its properties can change.

- AMH – automated mass handler - a system of motors, sensors and cams. The manufacturer confirmed a possible functionality in vacuum.

- LCM – laboratory condition monitor – it need not be in vacuum completely, but its sensors/probes should monitor conditions in the vacuum recipient – pressurized cylinder (IR sensor) and electronic balance (PT100) temperature and pressure in which the system runs. Temperature probes were found as applicable, but barometric probe LCM can measure only from 70 kPa to 130 kPa and does not enable measurement in vacuum. To measure the residual pressure, a different gauge was utilized – a Pirani vacuum gauge or an MKS Baratron. Moreover, the software controlling the gravimeter had a limitation not allowing the system to work outside the ambient pressure range from 70 kPa to 130 kPa. The system indicates non-measurable values in that case. This problem was eliminated in co-operation with the manufacturer programmer.

- Pressurized cylinder and flexible connecting capillary – these components withstand the vacuum without any problem. The difference of pressure is roughly 100 kPa and pressurized cylinder maximum filling pressure is 25 MPa, while it is filled only to 15 MPa, so it has a sufficient pressure reserve. Capillary operating pressure is 700 kPa but it can withstand up to 1 MPa, so a pressure reserve is sufficient, too.

- The Pt100 thermal sensors also work without problems in the vacuum.

3. VACUUM RECIPIENT GFS

After tests of the function of the whole system in the preliminary recipient, based on the experience during work, a new vacuum recipient was designed. We assumed that to demonstrate the gravimeter new parameters, many international comparisons will have to be accomplished, in

which it will be necessary to show a very low uncertainty of the standard. These comparisons have always a problem with a transfer standard, which does not exist for such low uncertainties. The only way how to compare it with other primary standard is a transportation of the whole gravimeter into the NMI, and to accomplish a direct comparison of both standards here. That is why we designed the vacuum recipient with an emphasis put on its transportability, see Fig. 4. And Fig. 5 and 6 show it in reality.

Fig. 4: Vacuum recipient for GFS

Fig. 5: Vacuum recipient GFS in the laboratory

Fig. 6: Opened vacuum recipient

It is a simple stainless steel cylinder with rubber sealing at its ends and welded-in vacuum fittings to connect pumping, measuring, electric and pneumatic connections. The cylinder has two strong duralumin ends to prevent its bending due to the atmospheric pressure. The lower cover furnished with legs and the upper cover is equipped with mechanism to lift and turn the upper cover. The recipient is pumped in vacuum mode by conventional dry vacuum pumps.

4. UNCERTAINTIES OF THE GFS IN THE ATMOSPHERIC MODE

The detailed uncertainty budgets for the atmospheric and vacuum modes, together with the resultant uncertainties are listed in Tab. 1 and 2.

Tab. 1. Uncertainty budget in atmospheric mode.

quantity		uncertainty (k = 1)	sensitivity coefficient	contribution to uncertainty
resolution	Δm	0.03 mg	1	0.03 mg
linearity	Δm	0.19 mg	1	0.19 mg
repeatability	Δm	0.24 mg	1	0.24 mg
correlated components of uncertainty weight	Δm_{cor}	0.006 mg	1	0.006 mg
weight of the tare weights	m_T	10 mg	$5 \cdot 10^{-6}$	0.00005 mg
density of tare weights	ρ_m	80 kg·m^{-3}	$1.25 \cdot 10^{-9}$	0.1 mg
volume of the cylinder	V_{ext}	$2.15 \cdot 10^{-5}$ m^3	0.041	0.88 mg
volume of accessories	V_{acc}	$1.42 \cdot 10^{-6}$ m^3	0.040	0.86 mg
coefficient of linear thermal expansion	α	$2.43 \cdot 10^{-6}$ K^{-1}	0.075	0.18 mg
temperature of cylinder	ΔT_{IR}	1.5 °C	$1.83 \cdot 10^{-7}$	0.27 mg
density of air	ρ_a	0.0026 kg·m^{-3}	$1.57 \cdot 10^{-6}$	0.004 mg
change in air density	$\Delta \rho_a$	0.00054 kg·m^{-3}	0.0025	1.37 mg
TOTAL (k = 1)	-	-	-	2 mg
sampling balance (k = 2)	-	-	-	0.0125[mg/(mg/s)]× ×Q_m[mg/s]
TOTAL (k = 2)	-	-	-	4 + 0.0125[mg/(mg/s)]× ×Q_m[mg/s]

Tab. 2. Uncertainty budget in vacuum mode.

quantity		uncertainty (k = 1)	sensitivity coefficient	contribution to the uncertainty
resolution	Δm	0.03 mg	1	0.03 mg
linearity	Δm	0.19 mg	1	0.19 mg
repeatability	Δm	0.07 mg	1	0.07 mg
correlated components of uncertainty weight	Δm_{cor}	0.006 mg	1	0.006 mg
weight of the tare weights	m_T	10 mg	$5 \cdot 10^{-6}$	0.00005 mg
density of tare weights	ρ_m	80 kg·m^{-3}	$1.25 \cdot 10^{-11}$	0.001 mg
volume of the cylinder	V_{ext}	$2.15 \cdot 10^{-5}$ m^{-3}	0.0003	0.0075 mg
volume of accessories	V_{acc}	$1.42 \cdot 10^{-6}$ m^{-3}	0.0004	0.0072 mg
coefficient of linear thermal expansion	α	$2.43 \cdot 10^{-6}$ K^{-1}	0.0005	0.0012 mg
temperature of cylinder	ΔT_{IR}	1.5 °C	$1.83 \cdot 10^{-9}$	0.0022 mg
density of air	ρ_a	0.0026 kg·m^{-3}	$1.57 \cdot 10^{-8}$	0.00004 mg
change in air density	$\Delta\rho_a$	0.00054 kg·m^{-3}	0.0003	0.00165 mg
TOTAL (k = 1)	-	-	-	0.31 mg
sampling balance (k = 2)	-	-	-	0.0125[mg/(mg/s)]× ×Q_m[mg/s]
TOTAL (k = 2)	-	-	-	0.62 + 0.0125[mg/(mg/s)]× ×Q_m[mg/s]

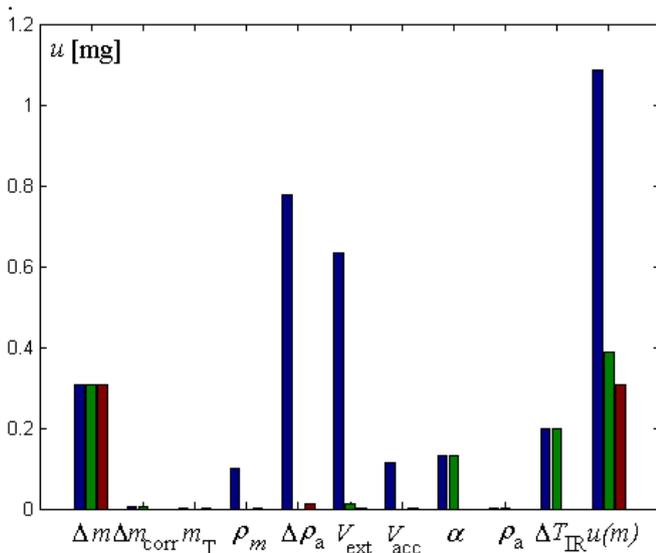Very similar table is also for weighing in a hermetic mode.

Fig. 7: Uncertainty of the amount of flow for 1.1 liter cylinder in various modes – atmospheric mode – blue, hermetic mode – green, vacuum mode – red

When comparing initial atmospheric weighing with the work in special closable box, then the use of hermetic mode significantly increases uncertainty of determination of the change in mass of the pressurized cylinder, see Fig. 7. As it is seen, vacuum mode is not such a significant advantage; however another issue must be also accounted for. This is a condensation of water on the cylinder surface during higher flows. In the vacuum mode, this issue will be practically eliminated.

However, a decrease of uncertainty of weighing will not reflect into the whole of applicable flows and especially of amounts of flow, due to the influence of the time measurement uncertainty, see Fig. 8.
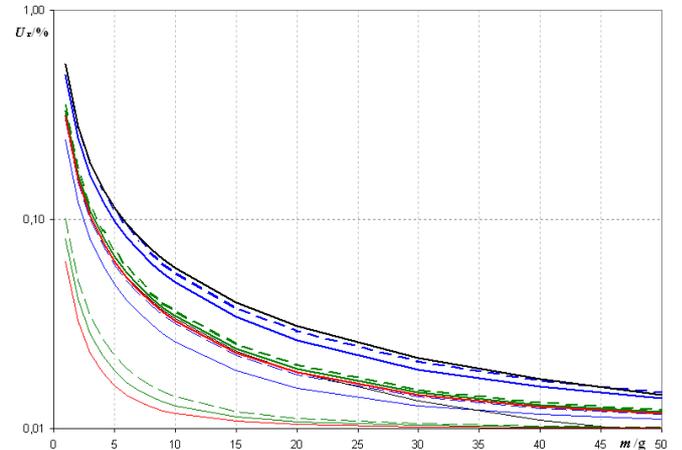

Fig. 8: Comparison of relative uncertainties of flow for various modes – 200 mg/s –thick line, 0,2 mg/s – thin line, DHI specifications – black, atmospheric mode – blue line, hermetic mode – green, vacuum mode – red line, 1.5 liter cylinder - broken line

While for small flows and smaller amounts flowing through, the uncertainty in hermetic mode decreases four times and in the vacuum mode decreases nearly five times, then for great amounts of flow the manufacturer declares the uncertainty lower by nearly 7 %. It should be taken into account that the manufacture's declaration is one thing and the uncertainty acknowledged by CMC is the other thing. Only a few national metrology laboratories all over the world can provide the conservative uncertainty level of 0.05 % (k = 2) from the measured value.

If we compare the initial atmospheric weighing with the work in special closable box, the solution is beneficial mainly for the secondary etalon Freon leak of large values, which usually have a bulky structure, see Fig. 9 and 10. However, it is necessary to realize that this solution was intended especially for them, since there exists a better solution for small and compact ones (Mettler comparator).

Fig. 9: Freon leak of large values

Fig. 10: Freon leak in a hermetic box with a camera for reading pressure of freon

5. RESULTS OF THE COMPARISON GFS IN VACUUM AND IN ATMOSPHERIC PRESSURE

Tab. 3. The measured difference between vacuum and atmospheric mode in calibration characteristics of the 500 sccm molbloc in % of measured value (MV).

Nominal flow – 500 sccm molbloc	Difference on molbloc with GFS in vacuum and atmospheric pressure	Scatter
[sccm]	[% from MV]	[% from MV]
500	0.016	0.008
400	-0.017	0.006
300	-0.026	0.002
200	-0.027	0.002
100	-0.025	0.009
50	0.012	0.009

6. CONCLUSIONS

Modification of the GFS using vacuum weighing improved the resultant expanded ($k = 2$) uncertainty of the national standard of low gas flow from 0.1 % to 0.05 % due to suppressing of all effects of uncertainty depending on the density of cylinder and the uncertainty of density of the ambient air. Also, now the condensation of water on the cylinder is quite impossible even when temperature drops below the dew point.

REFERENCES

[1] KRAJÍČEK, Z., PRAŽÁK, D., HAJDUK, T., TESAŘ, J.: Experiences with traceability of molbloc flowmeters to primary gravimetric mass flow standard. *Metrologie*. Vol. 56 (2009), No. 1, p. 38 - 43.
[2] KRAJÍČEK, Z., PRAŽÁK, D., VIČAR, M., TESAŘ, J., STANĚK, F.: Zajištění metrologie malého průtoku plynů – nový státní etalon dynamické gravitometrie – GFS. *Metrologie*. Vol. 19 (2011), No. 1. p. 18 – 23.
[3] KRAJÍČEK, Z.: *Rozšírenie rozsahu primárného etalónu malého hmotnostného prietoku plynu.* Bratislava: STU, 2013. ISBN 978-80-227-4014-2.
[4] ROMBOUTS, C., BAIR, M., BARBE, J., WRIGHT, J. D., KRAMER, R., KRAJICEK, Z.: A Comparison of Primary Gas Flow Standards Spanning the Range from 10 sccm N_2 to 10 slm N_2. *Measure* Vol. 9 (2014), No. 4, p. 46-54.