

CURRENT DESIGN TRENDS IN THE MODULES OF NATIONAL STANDARDS FOR THE UNITS OF LIQUID VOLUMETRIC FLOW RATE (VOLUME) WITHIN THE RANGE OF 10^{-5} to 10^3 mL/min

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UDC 006.3

The present article discusses the provision of metrological support for measuring liquid mass and volume in a flow, as well as liquid mass and volumetric flow rates within the microflow range of 10^{-5} to 10^3 mL/min, to satisfy needs of leading sectors of the world economy. Drawing on an analytical review, the main metrological and performance characteristics of national standards are presented. The article uses the dynamic weighing method to outline the basic principles of flow generation in national gravimetric and volumetric standards during liquid mass and volume measurements. The design and operating principles of key modules available in national standards are considered. Methods for supplying liquids to collection vessels and their design are described, taking into account the necessity to minimize liquid evaporation, as well as the impact of capillary force and buoyancy. The primary sources of uncertainty in measuring the mass and volume of liquid using the dynamic weighing method are considered, along with methods for minimizing such uncertainties. A modified model for the dynamic measurement of liquid mass flow rate is proposed, which takes the primary sources of uncertainty into account. The influence of various sources of uncertainty on the metrological characteristics of national standards is discussed.

Keywords: capillary forces, liquid evaporation, degassing, time drift, buoyancy, flow-rate stability.

Introduction. In 2010–2020, a consortium of foreign national research institutes, medical centers, and for-profit enterprises from Germany, Portugal, France, Czech Republic, Denmark, Switzerland, Great Britain, and the Netherlands significantly improved the metrological infrastructure for transferring units of liquid mass and volume in a flow, as well as liquid mass and volumetric flow rates within the flow range of 10^{-5} to 400 mL/min (henceforth referred to as the microflow range) [1].

The European Union is currently implementing a collaborative research project *Metrology for Drug Delivery II* funded under the European Metrology Research Program [1–3]. The project team includes nine national and research metrology institutes (IPQ, Portugal; CETIAT, France; CMI, Czech Republic; DTI, Denmark; METAS, Switzerland; NEL, UK; NQIS, Greece; RISE, Sweden; KRIS, Korea), four measuring instrument (MI) producers (DNV GL, Netherlands; HSG-IMIT, Germany; INESC MN, Portugal; BHT, Netherlands), as well as two university hospitals (THL, Germany; UMCU, Netherlands). The project aims to improve the accuracy of instruments for measuring a quantity of liquid and its flow rate within the microflow range in existing drug dosing devices, such as infusion pumps, as well as calorimetric, thermoanemometric, and Coriolis flowmeters. In order to achieve this aim, an expansion of the existing metrological infrastructure is planned.

The medical demand for providing metrological support for MIs operating within the microflow range is due to the need for accurate dosing of drugs to patients in neonatology (i.e., a field of medicine studying the development, diseases, and pathologies of newborns) [3], oncology, as well as other fields of medicine.

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Increased interest in instruments measuring liquid flow rates within the microflow range has been noted in various sectors of the Russian Federation:

in the pharmaceutical industry when preparing inert and reactive gas-liquid mixtures, as well as producing active pharmaceutical ingredients;

in medicine for representative multi-infusion systems and in the preparation of water for hemodialysis;

in the chemical industry when dosing toxic components used for polyvinyl chloride production; for the fractional separation of proteins, biopolymers, polymers, and nanoparticles;

in the food industry when dosing ultra-small quantities of food additives in the production of beer, milk, soft drinks, mayonnaise, and ketchup;

in the petrochemical industry when dosing neutralizers, demulsifiers, and corrosion inhibitors used in oil refining processes, as well as when measuring out additives to heavy fuel for marine engines;

in the research centers of the Russian Academy of Sciences and research universities when dosing methanol into a direct methanol fuel cell, as well as in the studies of optical fibers, rocket fuel, gas-vapor mixtures, and chemical reagents.

Over thirty types of MIs operating within the microflow range are registered in the Federal Information Fund for Ensuring the Uniformity of Measurements (FIFEUM). These MIs differ according to the underlying operating principle: Coriolis (mass) flowmeters, ultrasonic (volumetric) flowmeters, electromagnetic (volumetric) flowmeters, thermal (mass-calorimetric and thermoanemometric) flowmeters.

While being less common than other MI types, thermal flowmeters offer several advantages, such as no moving parts, a full-flow cross-section, as well as insensitivity to spatial arrangement and air transportation. The operating principle of thermal flowmeters is based on measuring the flow-rate-dependent thermal effect on the flow or body coming in contact with the flow [4]. The manufacturer recommends using this flowmeter within the liquid flow rate range of 0.08–16.70 mL/min [5].

A separate group of flowmeters is infusion pumps (not registered in the FIFEUM) are used in medicine for the invasive treatment of patients.

The arrival on the market of numerous MIs operating within the microflow range explains the need for improving the metrological infrastructure of the Russian Federation when transferring volumetric and mass units of liquid in a flow, as well as those of volumetric and mass flow rates within the flow range of $1.7 \cdot 10^{-2}$ –170 mL/min. This aim is to be fulfilled by extending the range of reproducible flow rates of GET 63-2019 *State Primary Special Standard of Units of Mass and Volume of Liquid in a Flow and of Mass and Volume Flow Rates of a Liquid* [6]. Furthermore, an improved base of measurement standards will allow MIs to be verified and calibrated across a practically significant range of liquid flow rates.

The present study aims to identify best practices in designing the modules of national measurement standards for the units of liquid volumetric flow rate (volume) within the microflow range, as well as in applying correction factors to the model for performing dynamic measurement of liquid flow rate (amount). The results will help to establish engineering guidelines for the development of key modules comprising the EU-4 standard unit, which is carried out in the context of GET 63-2019 improvement.

Design features of modules comprising national standards for the units of liquid volumetric flow rate (volume) within the range of 10^{-5} to 10^3 mL/min [3–18]. Reliable estimation of liquid flow rate and amount within the microflow range constitutes a challenging task since numerous sources of uncertainty, including those arising due to human factors, need to be controlled and managed. The implementation of optimal technical solutions inherent in the design of a primary measurement standard determines the level of its metrological characteristics.

Table 1 presents the metrological characteristics and liquid flow rate ranges of national measurement standards for the units of liquid flow rate and amount (henceforth referred to as measurement standards), demonstrating the current state of the metrological base [3–18], which reflects the needs of various sectors of the world economy. The following abbreviations are given in the table: FG – flow generators; DET – diaphragm expansion tank; SP – syringe pump; P – pump; CHT – constant head tank; GPC – gear pump having a flow controller; T , p , Q – temperature, pressure, and liquid flow rate, respectively; U_p – expanded measurement uncertainty.

An analytical review of experience in creating and operating measurement standards at national research and medical centers [1–18] resulted in the formulation of the main technical solutions for carrying out improvements to GET 63-2019,

TABLE 1. National Gravimetric and Volumetric Measurement Standards

Country, NMI, reference	FG	Q_V , mL/min	U_p , % ($k = 2$)	T , °C	p , MPa
France, LNE-CETIAT [3]	DET	$1.7 \cdot 10^{-2}$ – $1.67 \cdot 10^2$	0.10	10–50	0.10–1.00
Switzerland, METAS [7]	DET	$5 \cdot 10^{-5}$ – $4 \cdot 10^2$	0.07	22	0.10–1.00
Switzerland, METAS [7]	SP	$5 \cdot 10^{-5}$ – $4 \cdot 10^2$	0.90–0.07	22	0.10–1.00
Czech Republic, CMI [8]	P	10^{-3} – 10^2	0.20–0.05	25 ± 5	–
Czech Republic [9]	SP	10^{-3} – 10^2	0.60–0.05	25 ± 5	0.14–0.34
Denmark, DTI [10]	SP	$1.7 \cdot 10^{-5}$ – 10^2	4.00–0.05	–	0–5.00
USA, NIST [11]	CHT	10^{-4} –1	5.00–1.00	–	$1.7 \cdot 10^{-2}$
USA, NIST [12]	SP	10^{-4} –1	3.00–1.00	–	–
Portugal, IPQ [13]	SP	$2 \cdot 10^{-3}$ –10	2.50–0.30	20 ± 3	0–2.00
Indonesia, LIPI [14]	SP	1.51–19.6	11.60–1.60	–	–
Thailand, NIMT [15]	P	0.16–16.70	0.30	23 ± 2	–
Japan, NMIJ [16]	SP	$3.33 \cdot 10^{-1}$ – 10^3	0.10	20 ± 0.08	0–5.20
Netherlands, VSL [1]	GPC	$3.33 \cdot 10^{-1}$ –83.0	0.15–0.05	–	0–5.00
Bronkhorst [18]	GPC	$1.70 \cdot 10^{-2}$ –3.3	5.0–0.62	–	0–6.00

including the development of the EU-4 standard unit. This unit allows the measurement range of GET 63-2019 to be extended in terms of the liquid flow range of $1.7 \cdot 10^{-2}$ to 170 mL/min.

The operating principle of the examined national gravimetric and volumetric measurement standards consists in generating a liquid flow through the cross section of a MI under test, subsequently using the dynamic weighing method to determine the liquid mass (volume). After determining the liquid mass (volume) for the measurement interval τ , the mass (volumetric) flow rate of the liquid is calculated.

According to the review of design features, national measurement standards [3–18] include the following basic modules: a liquid storage and preparation module, a liquid flow generation module (flow generator), a flow regulation module, a measuring (work) table, a liquid mass measurement module, as well as a module for regulating environmental parameters.

The most commonly used liquid in national measurement standards, having well reproducible parameters, is previously prepared distilled water. The main advantages of distilled water include availability, lack of toxicity, low content of salts, trace elements, and minerals, as well as the concomitant possibility of transferring calibration and verification results to other liquids used for flow measurement in various industrial sectors [3].

Figure 1 shows the principle of measuring liquid amount and flow rate using a gravimetric measurement standard. In measurement standards equipped with flow generators, distilled water is used in a diaphragm expansion tank [3, 7], a pump [8], a constant head tank [11], and a gear pump having a flow controller [17–18]. In turn, flow generators in the form of syringe pumps employed in measurement standards [7, 9, 10, 12–14, 16] allow the use of various viscous liquids.

Liquid storage and preparation module. This module consists of a reservoir, filters, and a degassing device. It may also include a laboratory distiller, as well as a filtration, disinfection, degassing, and liquid temperature stabilization unit. Here, reservoir refers to a glass or metal (stainless steel) container designed to store liquid prepared for measurement.

The filters of the liquid storage and preparation system, which are located in the hydraulic path of a measurement standard, are used to remove particles from the liquid. This operation prevents clogging in pipeline sections and equipment installed in the hydraulic path of the measurement standard, the clear openings of whose capillary tubes have a small diameter. The inner diameter of some capillary tubes involved in the measurement process of individual measurement sections and equipment reaches 100 μm . Depending on the diameters of the hydraulic path of the measurement standard, it is possible to use filters having a pore size of 0.5 μm and above [3, 18].

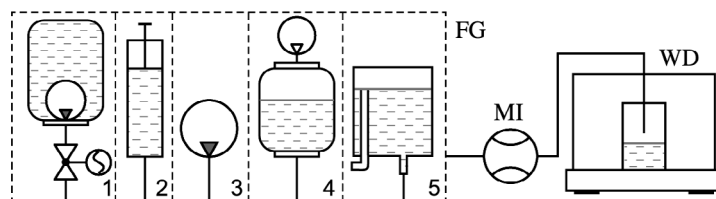


Fig. 1. Principle for measuring the units of liquid amount and flow rate using a gravimetric measurement standard: 1) gear pump having a flow controller, GPC; 2) syringe pump, SP; 3) pump, P; 4) diaphragm expansion tank, DET; 5) constant head tank, CHT; WD – weighing device; FG – flow generator; MI – measuring instrument.

The preliminary preparation of liquids used in advanced national measurement standards is a matter of priority [3, 7, 17]. In order to prevent the spread of bacteria, fungi, and algae in the liquid, the following measures can be taken: frequent liquid renewal (every two days) [18], use of a UV disinfection system [3, 7, 17], as well as the addition of fungicides and special chemicals to the liquid in small amounts [3].

In order to achieve high metrological characteristics of the measurement standard, it is necessary to determine parameters exhibited by the used liquid to ensure its stability. Therefore, for the purposes of assessing and controlling these influencing factors, it is recommended to regularly determine liquid parameters using laboratory methods.

Special attention is paid to stabilizing liquid temperature across the entire hydraulic path. To this end, the tanks of the measurement standard and hydraulic paths have to be set up under conditions enabling the maintenance of liquid temperature at a given level for a long period of time.

Pipeline segments in measuring sections and isolation valves must be completely filled with a moving liquid flow, with no air or gas bubbles present in the measuring section, since this can affect measurement accuracy. Such bubbles tend to agglomerate and clog the clear openings of some capillary tubes in individual measuring sections in the presence of stagnant zones having low local liquid flow velocity values, which occur due to sudden changes in the flow passage geometry in measuring sections. A gas bubble of significant dimensions can cause local pressure changes in capillary tubes, which in turn affects the liquid flow rate stability due to the compressibility of the gas phase. In addition, the passage of free gas through flow MIs typically has a significant impact on the reliability of their measurements. In order to solve this particular problem for reproduction within the low flow range, it is recommended to use degassers, transparent sections of the hydraulic path for visual control, isolation valves for draining air from the hydraulic path, and an operating algorithm that ensures the removal of free gas prior to measurements and controls its absence during measurement. For example, advanced national measurement standards [3, 7, 17] store preliminarily prepared water in stainless-steel tanks, in which vacuum pressure at room temperature is used to bring the liquid to boiling point to remove any dissolved gas from it.

Liquid flow generation module (flow generator). National measurement standards and verification units operating within a flow range greater than 167 mL/min generate liquid flow under forced pressure using pump units equipped with a variable frequency drive. The drawback of this liquid flow generator consists in the high- and low-frequency liquid flow pulsations produced by the rotating drive vanes of pump units. A significant liquid flow instability has a negative effect on the metrological performance of measurement standards within the liquid microflow range (<167 mL/min).

An analytical review of literary sources [3–18] shows that the liquid flow generator is implemented in national measurement standards using five different methods (see Fig. 1).

1. The gear pump in the diaphragm expansion tank maintains a constant value of liquid overpressure monitored by the corresponding sensor. Through opening or closing, the control equipment generates a stable flow rate characterized by low pulsation levels. [17, 18].

2. The liquid flow is generated using a driven syringe pump [7, 9, 10, 12–14, 16].

3. The liquid flow is generated by means of the pump H , creating a liquid pressure gradient in the pipeline.

4. Forced pressure liquid flow is created by overpressurized air (not exceeding 10^6 Pa) in the DET upper part displacing liquid from its bottom. A stable value of air overpressure generated by the compressor is monitored by an air overpressure sensor and a pressure reducer maintaining the specified air overpressure. This engineering solution minimizes fluctuations of the liquid flow [3, 7].

5. Forced pressure flow is generated due to the hydrometric liquid column in the constant head tank. The disadvantages of this method include a small overpressure in the pressure pipeline, which is determined by the height of the hydrometric liquid column [11]. Therefore, for a stable liquid flow rate, it is vital to ensure a constant pressure drop in the measuring pipeline section of the instrument under calibration (study) regardless of the local resistance by selecting the appropriate pipeline inner diameter of the measuring section [3].

Flow regulation module. The module comprises a piezoelectric control valve equipped with a flowmeter. Via the control function, the flowmeter adjusts the position of the piezoelectric control valve, ensuring that the liquid flow rate is maintained at a given value [7].

When flow rate values are reproduced by a measurement standard within a broad dynamic range, it is recommended to implement a supply manifold, i.e., several parallel capillary tubes having flow control systems that are assembled in separate measuring sections, each equipped with isolation valves.

The number of individual measuring sections is determined taking into account the overlap of their flow characteristics within a dynamic range of 10:1–100:1 [3, 11]. The pressure drop stability is also ensured by selecting capillary diameters that establish the laminar water flow regime in them [3]. Under laminar conditions, flow stability is strongly affected by liquid viscosity dependant on temperature. Therefore, in order to avoid changes in viscosity, the capillaries are immersed in a liquid-filled thermostatic bath, maintaining the liquid temperature in the capillary with a deviation of not more than 0.01°C [3].

It is recommended to use Coriolis and thermal flowmeters as standard flowmeters [18] due to their time-stable metrological characteristics, as well as high-frequency analog and digital output signals.

The hydraulic path design of the measurement standard must comply with the installation conditions of the used flowmeters as per instruction manuals: relative length of straight sections before and after the flowmeters; lack of mechanical stresses; coaxiality of connecting pipes; presence of earthing clamps, etc. In addition, it is recommended to use redundant isolation valves before and after the flow control system, enabling control of leaks between the valves and ensuring that the flow passages of flowmeters are filled with liquid at all times.

Measuring table of the measurement standard. The measuring (work) table of the measurement standard is designed for installing one or more MIs under study. Therefore, the length of the measuring table is determined by the number and types of MIs to be tested, provided that their installation requirements are met.

Since the liquid volume measured based on the readings of the MI under test and the measurement standard must be brought to the same conditions, it is necessary to measure the temperature and overpressure of the liquid near the tested MI, the liquid temperature in the collection vessel of the weighing device, as well as the atmospheric air pressure prior to and/or after determining the liquid volume.

The inlet and outlet of the measuring table should be equipped with isolation valves for shutting it off when installing and dismantling MIs, as well as an additional valve to release excess pressure. It is not recommended to equip the measuring table with a telescopic length compensator having a pneumatic (electric) drive, since this can lead to the emergence of stagnant zones that exhibit low local liquid flow rates, as well as the agglomeration of gas bubbles clogging the clear openings of capillary tubes in measuring sections. For convenience, it is recommended to place a tray under the measuring table to remove liquid when dismantling MIs.

Mass measurement module. The national measurement standards in [3–18] implement the method of dynamic weighing. A mass measurement system consists of weighing devices (balance) and collection vessels (beakers) for weighing liquid. The number of weighing devices is determined by the dynamic flow range of the measurement standard. For example, for standards operating within the range of 0.167 to 1.67 mL/min, the maximum mass of liquid in the measurement beaker, determined at a resolution of at least 10 µg, amounts to 5 g [3].

When collecting data, it is recommended to use the RS-232 (RS-485) interface for communication between the balance and the computer. This interface is used to match each obtained mass value with the corresponding time value, thus calculating the flow rate.

Weighing devices are placed on marble tabletops acting as platforms [3], with table supports set on an anti-vibration surface. In order to reduce the influence of free convection air currents on balance readings, a windscreen is installed for each weighing device [3–18]. In addition, balance operation may require the monitoring of ambient air parameters, i.e., the

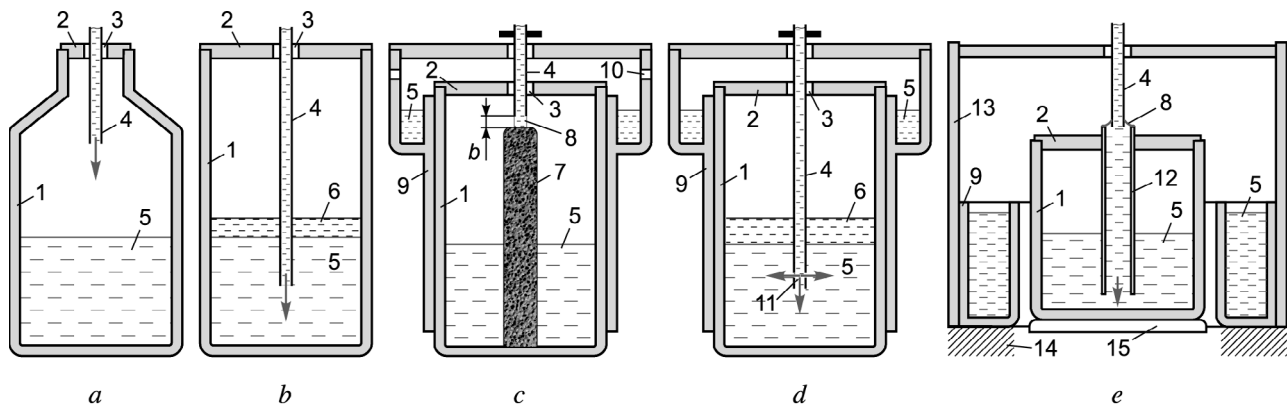


Fig. 2. Schematics of storage tanks according to [15], [1, 18], [7], [3], and [11, 12] (a–e, respectively): 1) beaker; 2) lid; 3) hole for the outlet capillary; 4) outlet capillary; 5) liquid (water or light oil); 6) oil layer; 7) fritted glass filter; 8) liquid (water or oil) bridge; 9) evaporation trap; 10) vent hole; 11) sprayer at the end of the capillary; 12) inlet capillary; 13) windscreen; 14) base of the balance; 15) weighing cell; *b* – gap between the end of the capillary and the glass filter.

maintenance of ambient humidity during operation and the adjustment of ambient temperature values within narrow ranges during the measurement period.

Another aspect of dynamic measurements involves accounting for air buoyancy when the collection vessel is being continuously filled with water. The buoyancy correction can be performed through the real-time adjustment of mass values when calculating the actual air and water densities [7].

Evaporation of liquid from the collection vessel. A significant problem associated with measurement standards operating within the low flow range consists in the evaporation of water from the collection vessel during measurement. In order to compensate for this effect, it is recommended to cover the collection vessel with a lid, use evaporation traps, or add a layer of high viscosity/lower density liquid over the liquid present in the collection vessel. Alternatively, these methods can be combined.

The NIMT measurement standard [15] employs a cylindrical collection vessel (beaker) 1 having a truncated cone top (Fig. 2a). In order to reduce the evaporation effect, the beaker 1 is covered with a lid 2. Water enters through an end of the outlet capillary 4 located above the water surface level 5. No data on the evaporation rate is provided. Experimental studies performed by the authors in [15] suggest that the evaporation rate ranged from 0.6 to 7 mL/min, depending on temperature and relative humidity.

The collection vessels of the VSL [1] and Bronkhorst [18] measurement standards are designed in the form of a cylindrical beaker (Fig. 2b) covered with a lid having a hole 3 for the outlet capillary. One end of the outlet capillary is submerged below the surface of water contained inside the beaker. Relative humidity inside the beaker reaches 98%, with condensate forming on its surfaces. The hole diameter is chosen to prevent condensate formation between the lid and the outlet capillary, as well as the emergence of a water bridge. The surface of water contained in the beaker is covered by an oil layer 6 [17], which significantly reduces the water evaporation rate down to $(0.02\text{--}0.53)\cdot 10^{-4}$ mL/min, depending on the volume of the beaker.

The design of the collection vessel in the METAS measurement standard [7] prevents water from dripping from the outlet capillary (Fig. 2c). To this end, a vertical fritted glass filter 7 is placed at the bottom of the beaker to form a 50 μm long water bridge 8 between its top and an end of the outlet capillary. The resulting capillary force in the glass filter sucks up water, preventing the formation of water droplets on its surface. Meanwhile, water in the evaporation trap 9 saturates the air with vapor. The degree of relative humidity is controlled by vents 10 positioned on the top of the evaporation trap. With the given design of the collection vessel, the water evaporation rate amounts to $(3.16 \pm 0.50)\cdot 10^{-6}$ mL/min, depending on the porosity of glass filters [7]. In this case, the evaporation rate value is an order of magnitude higher than when the water surface is covered with an oil layer. However, the capillary force remains virtually constant over time, having no effect on the relative increase in the mass of water in the collection vessel during measurements [7].

The collection beaker of the LNE-CETIAT measurement standard [3] is equipped with an evaporation trap in its upper part (Fig. 2d). One end of the outlet capillary is submerged below the surface of water contained in the beaker. A sprayer

II at the end of the outlet capillary decreases the impact of the submerged liquid jet flow on the weighing device. When using a layer of oil, the rate of water evaporation from the beaker is reduced to $1.5 \cdot 10^{-7}$ mL/min [3]. However, despite the reduced evaporation rate effect, an additional significant observed factor is related to the surface tension forces and buoyancy.

A distinctive feature in the collection beaker design of the NIST measurement standard [11, 12] is that the evaporation trap is independent of the beaker (Fig. 2e). The beaker is placed on the weighing cell *15*, with the evaporation trap set on the base of the balance *14*. A standard windscreen is removed from the base of the balance to cover the entire structure with a glass windscreen *13*, through which the outlet capillary is led to the beaker. An end of the outlet capillary (inner diameter – 0.5 mm) is introduced through the meniscus of the water bridge into the inlet capillary *12* (inner diameter – 1.5 mm). The water supply method is identical to that implemented in [7] using fritted glass filters, with the rate of water evaporation from the collection beaker reaching $4.1 \cdot 10^{-2}$ mL/min.

Consideration of capillary force and buoyancy. The theoretical models do not specify the mutual influence of the following considered aspects: influence of capillary force (surface tension force); liquid evaporation effect; drift of weighing devices; buoyancy depending on the liquid mass in the collection vessel with the outlet capillary immersed in liquid [3]. Experimental studies allow correction factors to be obtained depending on the immersion depth of the outlet capillary. For example, in [3], a 150 mm long capillary mounted on a vertical translation stage was immersed in water in fixed steps of 25 mm at a constant speed (proportional to the liquid flow rate). Following each immersion step, the capillary was held stationary for 30 min (proportional to the immersion time) in order to determine the mass of evaporated water. The experiments were performed for large (2000 g) and small (200 g) balances using correction factors of 2.4 ± 1.1 and 7.9 ± 1.1 g/m [3], respectively.

According to [12, 16], the proper positioning of the outlet capillary relative to the inlet capillary is controlled by a video camera capable of macro photography. For the sake of visual control over the positioning process, the outlet capillary *4* is designed in the form of a glass pipette (see Fig. 2e). For example, given an accurate positioning of the glass pipette (see Fig. 2e) relative to the inlet capillary in the NIST measurement standard [12], the surface tension force remains virtually constant over time, thus having no effect on the measurement of water mass in the collection beaker. The positioning of the glass pipette is carried out across three planes using micrometers mounted on a heavy-base tripod. The glass pipette and the inlet capillary must be completely filled with water. In this case, the water bridge provides a perfect (zero) contact angle θ with the pipette [12]. In the case of rupture of the water bridge, the surface tension force reaches $56 \cdot 10^{-3}$ N/m [11]. With inaccurate positioning of the glass pipette due to its incomplete wetting (drying), the surface tension force accounts for the emergence of noise (0.06 mg/min) during liquid mass measurement [11].

However, studies carried out in NMIJ [16] revealed that, when the outlet capillary (see Fig. 2e) measuring 1.76 mm in inner diameter was immersed in liquid (working liquid 5 – light oil having a density of $\rho = 818$ kg/m³), the upward capillary force E_c decreased the measured liquid mass value. The buoyancy of the outlet capillary E_{oc} attributable to liquid displacement and the mass of liquid M_{ws} present inside the outlet capillary below the water surface acted in opposite directions, thus increasing the measured liquid mass value. These forces caused a difference between the actual and measured liquid mass values $\Delta M_b = -E_c + E_{oc} + M_{ws}$. The ideal contact angle between the liquid (in this case, oil) bridge δ and the capillary depends on the material of the outlet capillary, the contact point position, and the properties exhibited by the liquid [16]. For example, the surface tension force for light oil amounts to $26 \cdot 10^{-3}$ N/m, while the contact angle reaches $\theta = 20^\circ$ [16]. At a light oil mass of 8.2 g in the collection vessel of the weighing device, a decrease in its mass value due to the capillary force E_c amounts to 25.3 mg. An increase in the immersion depth of the outlet capillary is found to increase the mass reading of the weighing device by 2 mg per 1 mm of immersion depth.

With the reproduction of liquid flow rates approaching 10^{-5} mL/min or below, an increased influence of the uncertainty source associated with the stick-slip phenomenon is observed. This phenomenon, which is characterized by the intermittent motion of liquid over the surface of the outlet capillary [7], is illustrated for different collection vessels when the outlet capillary is submerged below the water surface (see Fig. 2b, d) at a flow rate of 40 μ L/min [7]. The liquid (water or water-oil) slides intermittently over the surface of the outlet capillary, causing the reading of the weighing device to change by ± 10 μ L/min (noise). This phenomenon is attributed to the technical surface roughness of the outlet capillary, as well as the influence of the surface tension forces of the liquid. The stick-slip phenomenon can be minimized by using polished stainless steel, glass outlet capillaries [7], or capillaries having an oleophobic coating [10].

System for regulating environmental parameters. For a measurement standard to function properly, stable ambient temperature and humidity values must be maintained during its operation, with no significant impact of air currents on the measurement results of weighing modules. In order to meet these requirements, it is recommended to use designs preventing free and forced convection air currents in the room, including systems for cooling and heating, humidifying and drying the air, as well as MI for these parameters installed near the weighing modules of the measurement standard.

Sources of uncertainty. The analytical review revealed the following primary sources of uncertainty in the dynamic measurement model, affecting the metrological characteristics of the considered national measurement standards [3–18] (see Fig. 1):

calibration, time and temperature drifts, repeatability, linearity, and reproducibility of balance readings during mass measurement;

measurement of air and water densities; introduction of air buoyancy corrections;

buoyancy of the capillary and collection vessel;

dimensional changes of pipes and isolation valves;

measurement of time and synchronization intervals;

water evaporation from the collection vessel;

capillary force and surface tension coefficient of liquid in the outlet capillary and collection vessel;

temperature and pressure changes in the buffer volume between the MI and the end of the outlet capillary;

technical surface roughness of outlet capillaries [7];

flow rate stability [17];

presence or formation of air bubbles in the hydraulic system, as well as changes thereof over time [17];

reaction force at which water leaves the outlet capillary [3];

convective air currents [17].

The following sources make the greatest contribution to the uncertainty budget of the measurement standard: liquid evaporation; capillary force; liquid surface tension; technical surface roughness of the outlet capillary.

Measurement equation for the liquid mass flow rate. Reduced by the present authors to a general form according to the analytical review of national measurement standards [3–18], the modified model for performing dynamic measurement of liquid mass flow rate Q_M is presented in the form of the following equation

$$Q_M = \frac{1}{\tau_2 - \tau_1} \left\{ \left[M_2(1 - \rho_{a2} / \rho_w) + B_2 \right] - \left[M_1(1 - \rho_{a1} / \rho_w) + B_1 \right] + M_e \right\}, \quad (1)$$

where τ_1, τ_2 – initial and final values of the measurement time interval; M_1, M_2 – liquid mass at the initial and final measurement points; ρ_{a1}, ρ_{a2} – ambient air density at the initial and final measurement points; ρ_w – density of the material used for weights; B_1, B_2 – combined correction factor for the liquid mass values at the initial and final measurement points; M_e – mass of liquid evaporated during the measurement interval.

The combined correction factor for the liquid mass values at the initial and final measurement points is determined as follows:

$$B_{i=1,2} = \sum_{j=1}^5 k_{i,j}, \quad (2)$$

where $k_{i,j}$ – corrections for the capillary force $k_{i,1} = \pi d_{oc} \gamma \cos(\theta_i) / g$; air buoyancy $k_{i,2} = V_l \rho_{ai}$; buoyancy of the inlet capillary $k_{i,3} = \rho_{li} \Delta l F_{oc}$; buoyancy of the collection vessel $k_{i,4} = V_c \rho_{ai}$; buoyancy of outlet capillary submerged below the liquid surface in collection vessel $k_{i,5} = -V_{oci} \rho_{li}$; d_{oc} – inner diameter of the outlet capillary; γ – surface tension force; θ – contact (wetting) angle; g – gravitational acceleration; V_l – liquid volume; Δl – distance between the end of the outlet capillary and the water surface in the collection vessel; F_{oc} – cross-sectional area of the outlet capillary; V_c – volume of the collection vessel walls; V_{oc} – volume of the outlet capillary submerged below the water surface in the collection vessel.

The contribution of each correction $k_{i,j}$ from Eq. (2) varies according to individual situation depending on the liquid flow rate range and the design features of the collection vessel (see Fig. 2). For example, a fritted glass filter in the collection vessel [7] and a precise positioning of the outlet capillary relative to the inlet capillary [12] eliminate the capillary force effect.

TABLE 2. Uncertainty Sources in the Bronkhorst Standard

No.	Sources of uncertainty	$u(x_i)$, % ($k = 2$)	C, %
1	Calibration of the balance	$3.0 \cdot 10^{-5}$	0.08
2	Time drift of the balance	$1.0 \cdot 10^{-3}$	2.80
3	Balance resolution	$6.7 \cdot 10^{-5}$	0.18
4	Balance linearity	$6.7 \cdot 10^{-4}$	1.80
5	Balance repeatability	$8.0 \cdot 10^{-4}$	2.16
6	Temperature drift of the balance	$2.0 \cdot 10^{-4}$	0.54
7	Air density during calibration	$1.3 \cdot 10^{-3}$	3.38
8	Density of weights	$7.5 \cdot 10^{-4}$	2.03
9	Air density	$1.0 \cdot 10^{-2}$	27.92
10	Liquid density	$3.7 \cdot 10^{-2}$	10.04
11	Dimensional changes of the hydraulic path	$1.1 \cdot 10^{-3}$	2.88
12	Dimensional changes of isolation valves	$1.4 \cdot 10^{-3}$	3.78
13	Time measurement	$7.7 \cdot 10^{-1}$	8.69
14	Evaporation of the liquid	$2.4 \cdot 10^{-3}$	3.19
15	Capillary force	$2.0 \cdot 10^{-2}$	27.00
16	Volume expansion of liquid	$2.6 \cdot 10^{-3}$	3.54

TABLE 3. Uncertainty Sources in the NIST Standard

No.	Sources of uncertainty	$u(x_i)$, % ($k = 1$)	C, %
1	Liquid density	$5.0 \cdot 10^{-3}$	13.89
2	Time measurement	$2.0 \cdot 10^{-3}$	5.56
3	Evaporation of the liquid	$2.0 \cdot 10^{-3}$	5.56
4	Capillary force	0	0
5	Mass measurement taking air buoyancy into account	$3.0 \cdot 10^{-3}$	8.33
6	Buoyancy of the capillary	$8.0 \cdot 10^{-3}$	22.22
7	Flow rate stability and repeatability	$1.6 \cdot 10^{-2}$	44.44

For the DTI measurement standard [10], the primary contributors to measurement uncertainty at flow rates over 0.167 mL/min include the following uncertainty sources: corrections for the outlet capillary buoyancy $k_{i,5}$ and capillary force $k_{i,1}$ between the outlet capillary and two liquid layers (water-oil) in the collection vessel (see Fig. 2b). At lower flow rates, uncertainty in measuring liquid mass at flow rates below 0.167 mL/min and measurement uncertainty associated with the capillary buoyancy correction $k_{i,1}$ at flow rates below 0.005 mL/min (90% at 0.005 mL/min) predominate. Meanwhile, changes in the capillary force $k_{i,1}$ associated with a variation in the water surface level inside the collection vessel become insignificant (about 20 μ m). As the liquid flow rate decreases, the measurement uncertainty of the DTI standard [10] rises: 0.5% ($k = 2$) at 0.167 mL/min and 5 % ($k = 2$) at a minimum flow rate of $8.33 \cdot 10^{-3}$ mL/min. Similarly, the measurement uncertainty of the NIST measurement standard [12] at $k = 2$ is as follows: 0.035% at 0.1 mL/min; 0.07% at 10^{-2} mL/min; 0.45% at 10^{-3} mL/min; 0.45% at 10^{-4} mL/min.

Described by Newton's third law, the reaction force at which the liquid leaves the outlet capillary is predominant at higher flow rates [3]. However, it is difficult to estimate this uncertainty source [17].

Since the mass of the collection vessel greatly exceeds that of the accumulated liquid, the introduction of corrections for the collection vessel buoyancy is essential at very low flow rates [17].

Uncertainty sources and their values for the Bronkhorst measurement standard [18] at 0.25 mL/min and the NIST measurement standard [12] at 0.1 mL/min are presented in Tables 2 and 3, where $u(x_i)$ is measurement uncertainty and C is the contribution to the uncertainty budget. Tables 2 and 3 indicate a fundamental difference in determining and estimating uncertainty sources for the Bronkhorst [18] and NIST [12] national measurement standards, associated with the use of different correction factors in Eq. (1).

For example, the Bronkhorst measurement standard [18] is described in detail with respect to all the components of uncertainty sources due to measuring liquid mass with the help of a weighing device, air, and liquid densities (see Table 2, items 1–10), whereas only the resulting uncertainty source value associated with the liquid mass measurement is presented for the NIST measurement standard, taking air buoyancy into account (see Table 3, item 5) [12]. Items 14 and 3 of Tables 2 and 3, respectively, reflect the effect of liquid evaporation from the collection beaker on metrological characteristics for the Bronkhorst [18] and NIST [12] measurement standards, which helps to assess the effectiveness of design solutions used in these measurement standards while minimizing the evaporation effect. It is also worth noting the original design of the collection beaker in the NIST standard [12] (see Fig. 2e), which eliminates the capillary force effect (see Table 3, item 4). Conversely, the design of the collection beaker in the Bronkhorst standard [18] contributes to the capillary force significantly affecting the metrological characteristics of the measurement standard (see Table 2, item 15).

Regional comparisons of the national measurement standards for liquid flow units of DTI, IPQ, LNE-CETIAT, METAS, VSL, MIKES, Bronkhorst, and FH Lübeck (EURAMET.M.FF-S7) were conducted from 2012 to 2015 within the range of 0.002 to 3.3 mL/min.

Conclusion. In the presented analytical review examining the design features of modules that comprise national standards, a rational method of liquid flow generation was determined using a diaphragm expansion tank to minimize liquid flow rate pulsations and fluctuations. The proposed designs of collection vessels in the considered standards can be used to reduce the contribution of the evaporation effect, as well as the influence of capillary force and buoyancy. According to the study into the primary uncertainty sources of liquid mass and volume measurements performed via dynamic weighing, along with considered methods for minimizing these uncertainties, each uncertainty source is found to make individual contributions depending on the liquid flow range and the design features of the measurement standard. The proposed modified model for dynamic measurement of liquid mass flow rate takes into account the primary uncertainty sources. The effect of uncertainty sources on the metrological characteristics of national measurement standards is evaluated. The information from the analytical review conducted in May 2020 provided the basis for designing the EU-4 standard unit, which is implemented in the context of improving GET 63-2019 as part of the metrological infrastructure of the Russian Federation.

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