



# Proposal of a novel approach to reference instrument and procedure definition to measure hydrogen volume and volumetric flow in a legal metrology framework

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## ARTICLE INFO

### Keywords:

Legal metrology  
Hydrogen meters  
Traceability  
Reference meters  
Master meters

## ABSTRACT

International standards concerning hydrogen measurement in legal frameworks are still lacking. This study proposes a novel approach to instrument and procedure definition for the legal measurement of pure hydrogen volume and volumetric flow rate according to main international standards of metrology. A wet drum meter filled with water is chosen as reference meter. The selected reference instrumentation and procedures are replicable in other laboratories. Simulations and calculations demonstrate that hydrogen diffusivity in water have no influence on error estimations, while water evaporation is considered because it affects the volumetric flow rate measurement. The expanded uncertainty of the total metrological chain is less than 0.25%. This reference procedure allows to perform tests for error of indication of new meters both using hydrogen and other flowing gases with the same measurement principle, instrumentation and procedures, helping the development and certification processes of new products useful for the upcoming energy transition.

## 1. Introduction

The energy transition from fossil-fuel to carbon-free energy sources is one of the most impelling challenges that governments, institutes, industries, and populations have to deal with in the next years. The goal of limiting the global temperature increase below 1.5–2 °C is well established after the Paris Agreement [1]. Renewable energies are considered the fundamental solution to lower the global carbon footprint, in particular in the power generation sector. However, their inherent limits in terms of discontinuous production and storage capacity make renewables not always compatible with the electricity demand [2]. The decisive help could be provided by hydrogen, which has been proposed by many scientific experts, private companies and public authorities as a fundamental driver through this energy transition [3]. In fact, hydrogen can be exploited by power-to-gas routes using electricity from renewable sources and through methanation, which would combine carbon dioxide with hydrogen to produce a synthetic and renewable natural gas [4].

Then, the gas can be stored for later use to match electrical and thermal needs. Regarding distribution over long distances, hydrogen can be transported by different means, even via existing and new gas pipeline networks as a pure fluid or in a blend with natural gas. Finally, hydrogen can be used in established or in original applications within the industrial sectors as well as the domestic ones [2].

The option of transporting hydrogen as pure or blended gas through pipeline networks poses some matters with respect to gas grid regulation and standardization, as confirmed by Jaworski *et al.* [5]. The effect of the addition of hydrogen to natural gas measurement have been studied in different works, such as [6,7,8,9,10]. All the studies demonstrate that gas meters seem not to be affected significantly by hydrogen addition to natural gas up to about 10 to 30%. However, international standards concerning the legal measurement of natural gas flow and volume exist, whereas analogue standards for hydrogen are still lacking. Hence, few preliminary international projects have been working on proposing and proving new solutions of volume and volumetric flow meters for pure hydrogen, as the United Kingdom project “Hy4Heat” [11], or for blends

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<https://doi.org/10.1016/j.measurement.2022.111882>

Received 21 July 2022; Received in revised form 17 August 2022; Accepted 31 August 2022

Available online 6 September 2022

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Nomenclature			
<b>Acronyms</b>			
1D	One-dimension	$r$	Diffusing particles hydrodynamic radius, (m)
OIML	International organization of legal metrology	$T$	Temperature, (K)
MID	Measuring instrument directive	$t$	Time, (s)
MPE	Maximum permissible error	$U$	Expanded uncertainty
MUT	Meter under test	$u$	Standard uncertainty
WDM	Wet drum meter	$V$	Volume, (m <sup>3</sup> )
		$v$	Velocity, (m/s)
		$x$	Spatial coordinate, (-)
		$y$	Molar fraction, (mol/mol)
<b>Symbols</b>		<b>Superscripts</b>	
$\alpha$	Advection constant, (-)	*	Quantity corrected
$\varepsilon$	Measurement bias, (-)	'	Quantity relative to the calibration-part procedure
$\lambda$	Diffusion constant, (-)	“	Quantity relative to certification- and verification-part procedure
$\mu$	Viscosity, (Pa • s)	sat	Saturation
$C$	Hydrogen concentration, ( $I_{H_2}/I_{H_2O}$ )	<b>Subscripts</b>	
$\bar{C}$	Mean hydrogen concentration, ( $I_{H_2}/I_{H_2O}$ )	avg	Average
$D$	Diffusion coefficient, (m <sup>2</sup> /s)	BLL	Bell prover
$E$	Volumetric flow rate percentage error, (%)	diff	Diffusion
$E_n$	Test bench comparison quantity, (%)	eva	Evaporation
$J$	Number of control volumes, (-)	H <sub>2</sub> O	water
$j$	Spatial index, (-)	in	Wet drum meter inlet
$K$	Coverage factor, (-)	out	Wet drum meter outlet
$k$	Boltzmann constant, (J/K)	min	Minimum
$N$	Number of time steps, (-)	max	Maximum
$n$	Time index, (-)	MUT	Meter under test inlet conditions
$\dot{n}$	Molar flow rate, (mol/s)	nom	Nominal
$n_{rev}$	Number of revolutions, (-)	ref	Reference meter
$P$	Pressure, (Pa)	WDM	Wet drum meter average conditions
$Q$	Volumetric flow rate, (l/h)		
$R$	Universal gas constant, (J/(mol K))		

with natural gas, as the EMPIR EURAMET project “NewGasMet” [12]. Nevertheless, it is becoming ever more pressing to select the instrumentation and to define the procedures for the metrological characterization and verification of new domestic gas meters suitable to measure hydrogen for gas trades with the same accuracy and uncertainty of natural gas measurements.

European projects as MetHyinfra [13] and MetroHyVe 2 [14] recently started, but they have not provided yet results on metrological activities and reference systems. New test rigs have been developed in Europe and used to test industrial and domestic gas meters [15,16,17], facing the challenges of reproducing the same quality level of natural gas metrological chains also using hydrogen blends. Also new facilities for testing of meters with pure hydrogen are under development or characterization in terms of metrological chain. TÜV-SUD facility is using a sonic nozzles reference system with good results in terms of a measurement uncertainty that is equal to 0.3% (with 95% of confidence) [18]. The reference systems selected in these new facilities are mainly sonic nozzles, turbine or Coriolis meters considering the volumetric flow rate as the main quantity of interest. Traditionally, though, gas meters are verified using volume reference systems (bell prover, piston prover) being tools for measurement of gas volumes. A reference system able to consider both volume and volumetric flow also with pure hydrogen is still lacking and could help the development and diffusion of new hydrogen meters.

Recommendations and standards are used as a guideline for the definition of the reference instrument selection and procedure described in this work. Among recommendations, OIML R 137 [19] and OIML R 140 [20] are provided by the International Organization of Legal Metrology that harmonize standards of gas meters and measuring systems for gaseous fuel at the international level. The application of these recommendations in the present work is a necessary step to make the

proposed procedure compatible with all metrological infrastructures worldwide. In the European legislation, the Blue Guide [21] ensures the broadest possible common understanding on implementation of the New Legislative Framework for the marketing of products. Furthermore, the European Regulation (EC) 765/2008 [22] and the Measuring Instruments Directive (MID) 2014/32/EU [23] are considered in this work. In particular, the Regulation 765/2008 is necessary for the compliance with the requirements for accreditation and market surveillance relating to the commercialization of products, whereas the Directive has the scope to “harmonize the laws of the Member States of the European Union relating to the making available on the market of measuring instruments”. Among the technical standards, the European standards about gas meters, *i.e.* EN 1359 (diaphragm meters), EN 14236 (ultrasonic meters), EN 17526 (thermal mass meters), EN 12261 (turbine meters), EN 12480 (rotary displacement meter) and EN 12405 (conversion devices), are observed in the present work to adopt the harmonized directives of the European Union member States relating to the homologation and the test requirements. The main issue in these technical standards is that hydrogen is not covered in their scope. Moreover, European standards as ISO/IEC 17025 (general requirements for the competence of testing and calibration laboratories) and ISO/IEC standard about conformity assessment, *i.e.* 17065 (requirements for bodies certifying products, processes and services), 17020 (requirements for the operation of various types of bodies performing inspection), 17021 (requirements for bodies providing audit and certification of management systems), 17043 (general requirements for proficiency testing), 17011 (requirements for accreditation bodies accrediting conformity assessment bodies), and 17050 (supplier’s declaration of conformity), are observed in compliance with the requirements for accreditation, inspection and notified bodies in terms of conformity, certification and tests.

The goal of this work is to propose a novel approach to the selection of the reference instrumentation and the definition of the procedures for the legal measurement of volume of pure hydrogen in agreement with the main international standards of metrology. These instructions aim at performing tests as a support to type approval evaluation of these new hydrogen domestic gas meters compliantly with the current certification processes to help the development and certification processes of new products useful for the upcoming energy transition, focusing the attention on performance tests for meter accuracy evaluation. Because other proposals are currently appearing in the metrology field, the procedure in this work represents one more solution than the existing ones presented in the previous paragraphs [13,14,15,16,17,18], which are still little and limited to a few laboratories without yet a full-blown global consensus.

The article is structured as follows. Section 2 deals with the instrument selection and procedure description comprising the calibration-part, certification-part and validation-part. Additionally, the measurement uncertainty calculation is described. Section 3 shows the results of the tests obtained by the applied measurement procedure. Finally, Section 4 discusses the results of the whole analysis and draws the final conclusions. As a support to the measurement procedure, Appendix A demonstrates that the diffusivity of the flowing gas in the liquid of the wet drum meter is negligible.

## 2. Materials and methods

This section details the selected instrument installed on the test bench and defines the procedure for its use. First, the principle at the base of the proposed procedure is depicted. Secondly, the choice of wet drum meters as reference meters is motivated. Thirdly, the measurement procedure is described in its three parts: calibration, certification and validation. Then, the test bench employed in this work, in addition to the reference meter, are described. Lastly, the measurement uncertainty calculation is illustrated.

### 2.1. Procedure principle

For sake of type approval evaluation, gas meter performances have to be compared to that of a reference meter placed in series. Specifically, error curves comparing the volumetric flow rate of the gas meter under certification, *i.e.* the so-called meter under test (MUT), with respect to the reference systems are determined at different volumetric flow rates to determine the MUT error curves. At each flow rate, the measurement error,  $E$  (%), is calculated as follows:

$$E = \frac{Q_{MUT}^* - Q_{ref}^*}{Q_{ref}^*} \cdot 100 \quad (1)$$

where  $Q_{MUT}^*$  and  $Q_{ref}^*$  are the corrected volumetric flow rate of the meter under test and the corrected volumetric flow rate of the reference, whose calculation is described in the following sections considering also water evaporation in the wet drum meter.

In general, the tests for type approval evaluation of gas meters should be performed by reference meters as well as systems and procedures able to guarantee traceability to an international reference standard. The selected reference meter must be stable in time. Furthermore, its performances must not depend on the nature of the flowing gas both in terms of chemical and mechanical stresses, as well as of impact on accuracy. The test equipment should give a certain metrological stability, resolution, repeatability, and uncertainty. Specifically, the uncertainty for type testing must be lower than 1/5 of the maximum permissible error (MPE) [19].

Therefore, the following sections describe in sequence the reference meter, the procedure and the instrumentation that are selected to guarantee the requirements of international reference standards. Furthermore, the method used for the calculation of the measurement

uncertainty is depicted.

### 2.2. Reference meter selection and evaporation model

The measuring systems employed for certification of gas meters are classified based on the gas volume passed or the gas flow rate measured, either mass flow rate or volumetric flow rate. On the one hand, measuring the flow rate has the advantage of reducing the experimental time for the tests, limiting the risks in laboratories when flammable gases are used because measuring average flow rate requires lower gas volumes. In contrast, measuring volume has the disadvantage that the duration of test is directly proportional to the meter resolution, hence in the case of low resolutions longer test duration collecting greater volumes are necessary to decrease the impact of the resolution on the total uncertainty. On the other hand, when the mass flow rate or volumetric flow rate are measured, it is necessary to integrate it over time to obtain volume, and measurement errors must be calculated as defined in the Measuring Instruments Directive 2014/32/UE Annex IV. Time integrations implicate synchronization issues with the flow oscillation during measurements and introduce another source of experimental errors. In contrast, when the volume is measured, such measurement errors are not encountered.

Several measuring systems are employed for certification and verification of gas meters, such as sonic nozzles, thermal mass flow meter and bell prover to name the most common, as seen previously. Cascetta *et al.* [24] classifies natural gas metering technologies in volumetric, non-volumetric and conversion devices. Sonic nozzle systems measure the mass flow rate of a flowing gas with a good accuracy and repeatability. Their measuring principle is based on the Venturi effect in a critical regime [25], where the mass flow rate is proportional to the inlet total pressure, while it is independent on the outlet pressure. Thermal mass flow meters measure the mass flow rate by means of the heat convection from a heated surface to the flowing fluid. Their measuring principle is based on King's law [26]. Both sonic nozzles and thermal mass flow meters are mature technologies for mass flow rate reference systems. In these cases, though, the conversion to volumetric flow rate must be computed by means of the calculated gas density and, for this reason, it highly depends on the gas characteristics. The gas meter calibration process is crucial for achieving good reference systems and may become complicated when the complexity and variety of gas mixing increase, or in presence of changes of composition. This aspect is particularly critical in the case of sonic nozzles and thermal mass meters. In their turn, Coriolis meters are not considered in this work due to their high costs and high uncertainty at the low pressures and gas densities at the conditions of interest of this work.

Because of the cited drawbacks of the mass flow rate measurement, the use of an inherent volumetric flow rate measurement principle is adopted in this study, where volume is the metrological quantity that has legal and economical relevance, as declared by MID 2014/32/UE Annex IV. The most used volume measuring system for test and calibration is the bell prover. Its measuring principle is based on the displacement of an air volume over time. Bell provers have a very good accuracy, repeatability, and stability in time, and they also have the main advantage of measuring directly the volume of the flowing gas in actual conditions. However, despite the several advantages of bell provers, another reference system with the same advantages of the bell prover is defined. This is done also to explore new ranges of calibration technologies and procedures to be compared to other proposed solutions. The wet drum gas meter (WDM) is selected in this study because they are more accessible instruments with respect to bell provers both in terms of dimensions, costs and safety with flammable gases. For this reason, they allow to ease replicability and comparison among different laboratories. Additionally, the wet drum meter can be defined as an instrument with an infinite capacity, thus continuous refilling with large hydrogen volumes is not needed as it is in a bell prover. Finally, risk assessments in the laboratory can be highly eased by the utilization of

wet drum meters compared to bell provers due to the smaller volumes that allow to ease inertization for safety issues and hydrogen discharge in case of security alarms. As a conclusion, the selection of the wet drum meter is made to make the procedure the most replicable in terms of both costs and dimensions.

Wet drum meters work on the principle of a positive displacement due to the flowing gas [27], so the volume is measured directly for all gases and gas mixtures with equal accuracy. These meters have an internal measuring drum partly submerged in packing liquid, for instance water or low viscosity oil, and rotated by the gas flow pressing over vanes that identify rigid measuring chambers. These chambers are filled and emptied periodically in this way. The measuring drum is coupled to a needle-dial and cumulating counter to measure the gas volume, while the flow rate can be measured with an impulse-counter and a high-resolution timer.

In calibration and research laboratories, especially where the conformity assessment process as notified bodies is active, the most preferred used medium is oil. Oil is usually preferred because the evaporation problems are limited to a minimum and oil does not cause corrosion of the measuring system components, and thus does not introduce additional components of measurement uncertainty. Oil is mainly used when the tested gas is air, as for example in bell provers, where oil acts as sealing liquid between a vertical inner tank and the outer shell. When the tested gas is a flammable gas, safety issues must be taken into account.

Considering the case of tests with natural gas, more than hundreds of liters of gas could pass through the bell prover and the risk of combustion increases considerably. Indeed, notified body prefers to use other test benches for meter verification with natural gas, where the reference meter (as for example, meter with an inner constant volume, as a diaphragm meter or a wet drum meter) does not need to be filled with oil necessarily, reducing the risk of combustion totally. The volume of a drum meter is bigger than tens of liters, and oil can act as combustible in case of fire in the laboratory. Also, Park *et al.* [28] underline the advantages of water with respect to oil.

Testing meters with hydrogen has the same issues and risks related to use of oil of testing meter with natural gas. In this paper, the main aim is to adopt test benches similar to that used with natural gas also for hydrogen and to propose a reference system and procedures that could be easily replicable in other laboratories (notified body, institutions, meter manufacturers) keeping as lower as possible the risk and the costs of the system. Moreover, oil utilization may lead to different disadvantages in terms of cost and replicability being more expensive than distilled water and not being unique. Indeed, the choice of oil could restrict the validity and replication of this work depending on the type of chosen oil. Finally, no literature studies deal with hydrogen diffusivity in oil, that should be evaluated for hydrogen utilization in drum meters. For all the above-mentioned reasons, water can be a better option than oil.

Due to the presence of water in the meter, the effect of gas diffusivity in water and water evaporation must be analyzed. The former phenomenon is neglected because its effects are negligible, as demonstrated beyond in Appendix A. On the other hand, the latter is considered in all the measurement procedure parts, as described in the following sections. In fact, a portion of the water of the wet drum meter evaporates into the gas flowing in the wet drum meter itself. Simultaneously, the inlet gas humidity affects the amount of evaporated water. Because of this phenomenon, the measurement of the outlet volume from the wet drum meter comprises both the flowing gas and the evaporated water volume. Hence, the effect of water evaporation shall be considered to correct the WDM flow measurement prior to comparing it with the MUT measurement. If this is not done, the measurement would be characterized by errors even in the order of percent points, that are not acceptable in terms of metrological traceability. In 2002 Park *et al.* [28] defined an experimental set up to account for water evaporation in wet drum meters. The procedure is very practical, but it requires the utilization of a

bell prover. Because the bell prover cannot be used for flammable gases as hydrogen, as anticipated in the previous section, the following paragraph shows how to compute the evaporated water volumetric flow rate correctly for any gas flowing through the wet drum meter.

The evaporated water volumetric flow rate in the wet drum meter,  $Q_{eva}$  (l/s), is calculated as follows. First, the inlet and outlet water mole fractions in the flowing gas,  $y_{H_2O,i}$  (mol<sub>H<sub>2</sub>O</sub>/mol<sub>gas</sub>), and the inlet and outlet water molar flow rates,  $\dot{n}_{H_2O,i}$  (mol<sub>H<sub>2</sub>O</sub>/s), are calculated via Raoult's Law as [29]:

$$y_{H_2O,i} = \frac{RH_i P_{H_2O}^{sat}(T_i)}{P_i} \quad (2)$$

$$\dot{n}_{H_2O,i} = \dot{n}_{tot,i} \bullet y_{H_2O,i} \quad (3)$$

where  $RH_i$  (%) is the measured relative humidity of the flowing gas at section  $i$  of the wet drum meter,  $P_{H_2O}^{sat}(T_i)$  (Pa) the water saturation pressure as a function of the measured temperature of the flowing gas,  $P_i$  (Pa) the measured pressure of the flowing gas, and  $\dot{n}_{tot,i}$  (mol/s) are the total molar flow rates. The water saturation pressure as function of temperature can be calculated by the formulations provided by the "International Association for the Properties of Water and Steam" (IAPWS) [30]. In its turn, the total molar flow rates can be calculated by the ideal gas model starting from the volumetric flow rate of the flowing gas. The subscript  $i$  indicates either the inlet or outlet section of the wet drum meter referred to as *in* and *out*, respectively.

Next, the evaporated water flow rate in the wet drum,  $\dot{n}_{eva}$  (mol/s), and the corresponding evaporated water volumetric flow rate,  $Q_{eva}$  (m<sup>3</sup>/s), are computed as follows:

$$\dot{n}_{eva} = \dot{n}_{H_2O,out} - \dot{n}_{H_2O,in} \quad (4)$$

$$Q_{eva} = \frac{\dot{n}_{eva} R T_{WDM}}{P_{WDM}} \quad (5)$$

where  $R$  (J/(mol K)) is the universal gas constant, while  $T_{WDM}$  (K) and  $P_{WDM}$  (Pa) the temperature and pressure averages between the inlet and outlet wet drum meter measured conditions, respectively. In particular, Eq. (5) is based on the assumption that vaporized water is modelled as an ideal gas in agreement with Raoult's Law, which is a very well-suited approximation for low pressure conditions, as in the case of domestic gas meter application. In fact, saturated steam behaves as an ideal gas for pressure values approximately even up to 1.1 MPa, which is a limit well above the partial pressure of water at the inlet and outlet of the wet drum meter [31]. At the room temperature of 20 °C, for instance, saturated steam compressibility factor is equal to 0.99865, proving the applicability of the ideal gas model.

### 2.3. Three-part procedure definition

This section deals with the description of the procedure proposed in this paper for the performance evaluation tests of new gas meters. As explained earlier, the procedure is divided in three parts: calibration, certification and validation. During the calibration-part, the wet drum meter is placed downstream of the bell prover and the tests are performed in air. During the certification-part, the wet drum meter is placed downstream of the meter under test and the tests are performed in hydrogen. During the validation-part, both the bell prover and the wet drum meter are used, and their volumetric flow rates are compared to that of a travelling measurement standard; in this part, tests are performed both in helium and air. At last, some remarks regarding the procedure are highlighted.

#### 2.3.1. Calibration-part procedure

Before depicting the error curves between the meter under test and the reference meter, which is the wet drum meter, the latter must be calibrated. The calibration of the wet drum meter consists in the

determination of its geometric volume. This volume is the empty space fillable with the flowing gas, therefore it is constant for each test as far as the liquid water free surface does not change appreciably due to evaporation and, importantly, it is independent on the gas itself. In this work, calibration is performed in air as required by the reference standards. The geometric volume can be calculated as the sum of the evaporated water and the inlet gas volume filling the wet drum during a revolution of the wet drum meter. The former is obtained via Eq. (5) from the measurements of pressure, temperature and relative humidity both at the inlet and the outlet of the wet drum meter. The latter is considered equal to the volume displaced by a well assessed reference system, such as the bell prover, placed upstream the wet drum meter. In fact, the bell prover is not affected by water evaporation and can be used with air safely in a laboratory. The number of revolutions and the duration of each test are also needed to shift from volumes to volumetric flow rates. The following section describes the model to calculate the wet drum meter geometric volume, where the quantities are indicated by a single quote mark to refer explicitly to this calibration-part procedure.

The geometric volume of the wet drum meter,  $V_{WDM}$  (l), is determined during calibration as follows:

$$V_{WDM} = \frac{Q'_{WDM}}{n'_{rev}} \cdot t' \quad (6)$$

where  $t'$  (s) and  $n'_{rev}$  (-) are the measured time duration and number of revolutions of each test during calibration, respectively.  $Q'_{WDM}$  (l/s) is the volumetric flow rate of the wet drum meter. It comprises two terms, the corrected volumetric flow rate of gas flowing into the wet drum meter,  $Q'_{WDM}^*$  (l/s), and the evaporated volumetric flow rate in the wet drum meter from Eq. (5),  $Q'_{eva}$  (l/s), as follows:

$$Q'_{WDM} = Q'_{WDM}^* + Q'_{eva} \quad (7)$$

Specifically, during calibration  $Q'_{WDM}^*$  (l/s) is equal to the gas volumetric flow rate measured by the bell prover,  $Q'_{BLL}$  (l/s), corrected to the wet drum temperature and pressure conditions as follows:

$$Q'_{WDM}^* = \frac{P'_{BLL} T'_{WDM}}{T'_{BLL} P'_{WDM}} Q'_{BLL} \quad (8)$$

where  $T'_{BLL}$  (K) and  $P'_{BLL}$  (Pa) are the measured temperature and the pressure of the bell prover during calibration.

The authors suggest performing calibration tests at different bell prover volumetric flow rates and calculating the geometric volume as the average of the geometric volumes calculated in all these different tests.

### 2.3.2. Certification-part procedure

The geometric volume determined from Eq. (6) during calibration can be used to calculate the corrected gas volumetric flow rate of the wet drum meter. This calculation can be performed for any test after the wet drum meter has been calibrated, i.e. during certification as well as validation. Also in this case, the effect of water evaporation shall be considered by Eq. (5). The corrected volumetric flow rate is then used as the reference volumetric flow rate to calculate the measurement error by Eq. (1), where *ref* is substituted by *WDM*. The following section describes the model to certificate meters under test, where quantities are indicated by a double quote mark to refer explicitly to this certification-part procedure.

First, Eq. (7) is rearranged to obtain the corrected wet drum meter volumetric flow rate during certification,  $Q''_{WDM}^*$  (l/s), as follows:

$$Q''_{WDM}^* = Q''_{WDM} - Q''_{eva} \quad (9)$$

where  $Q''_{eva}$  (l/s) is the evaporated volumetric flow rate in the wet drum meter during each test by using Eq. (5). In its turn, the wet drum

volumetric flow rate,  $Q''_{WDM}$  (l/s), is computed starting from the geometric volume determined during calibration as follows:

$$Q''_{WDM} = \frac{V_{WDM}}{t''} \cdot n''_{rev} \quad (10)$$

where  $t''$  (s) and  $n''_{rev}$  (-) are the measured time duration and number of revolutions of each test during certification, respectively.

Finally, it is possible to compare the corrected WDM volumetric flow rate with that of the MUT by Eq. (1), where the volumetric flow rate indicated by the meter under test during certification,  $Q''_{MUT}$ , is corrected to the same thermodynamic conditions of the WDM. Specifically,  $Q''_{MUT}$  is corrected to  $Q''_{MUT}^*$  applying the ideal gas model as follows:

$$Q''_{MUT}^* = \frac{P''_{MUT} T''_{WDM}}{T''_{MUT} P''_{WDM}} \cdot Q''_{MUT} \quad (11)$$

where the compressibility effects are not considered because the compressibility factor for hydrogen is almost equal to 1 at operative and standard conditions.

### 2.3.3. Validation-part procedure

The validation-part aims at verifying that the calibration of the wet drum meter is performed successfully. The authors suggest performing the validation-part also to acquire experience on the proposed procedure. However, this third part is not intended to be mandatory, but it can be performed voluntarily at other laboratories after the calibration-part. The validation-part consists in the direct comparison of the error curves of a travelling measurement standard with respect to both the bell prover and the wet drum meter, as described by ISO 17043:2010 standard "Conformity assessment – General requirement for proficiency testing". The following section describes the model to validate the calibration. Here the quantities are indicated again by a double quote mark as in the certification-procedure because calculations for both certification and validation are executed in the same manner.

Similarly, to the certification-part, Eq. (1) will be applied using both the bell prover and the WDM, while the MUT is substituted by a travelling measurement standard. Moreover, Eqs. (9) to (11) will be used to calculate the corrected wet drum meter and meter under test volumetric flow rate. In its turn, the bell prover corrected volumetric flow rate is calculated similarly to Eq. (8) as follows:

$$Q''_{BLL}^* = \frac{P''_{BLL} T''_{WDM}}{T''_{BLL} P''_{WDM}} \cdot Q''_{BLL} \quad (12)$$

where the compressibility effects are not considered because the compressibility factor for hydrogen is almost equal to 1 at operative and standard conditions.

In this study both air and helium are utilized in the validation-part at different flow rates, while a diaphragm meter is employed as travelling measurement standard. Air is used because it is the typical calibration gas for legal measurements. Helium is selected as the second validation gas because it is an inert gas with a low density similarly to hydrogen. Thus, the potential effects of the utilization of a low density gas, as well as potential leakage issues, are evaluated in this part. Both gases are used to demonstrate that the calibration of the wet drum meter is applicable to gases of any nature.

Once the error curves of the travelling measurement standard with respect to the bell prover and the wet meter are depicted for air and helium, the trends of the error curves of the two gases are compared. For low helium volumetric flow rates, the errors of the travelling measurement standard with respect to the bell prover and wet drum meter are expected to increase due to the mentioned effect of low gas density on diaphragm gas meters.

### 2.3.4. Procedure remarks

This section presents three additional remarks of the proposed

**Table 1**

Main characteristics of the reference meter employed in the procedure and of the meter under test, employed for sake of example in this work, including the information about manufacturer, type, as well as maximum and minimum volumetric flow rates,  $Q_{max}$  and  $Q_{min}$  respectively.

Gas Meter	Manufacturer	Type	$Q_{max}$ (m <sup>3</sup> /h)	$Q_{min}$ (m <sup>3</sup> /h)
Reference meter	Elster	NB 15	300	0.100
Meter under test	Bessel	G 4–10	10	0.040

procedure that can be observed by laboratories and may be considered during the type approval evaluation of new gas meters.

First, as explained, the calibration of the wet drum meter consists in the determination of the wet drum meter geometric volume, *i.e.* the empty space fillable with gas. This volume is constant for each test as far as the liquid water free surface does not change appreciably due to evaporation. In fact, water evaporation decreases the water volume inside the wet drum meter, increasing the actual geometric volume of the reference meter previously calibrated. Therefore, it is suggested controlling the liquid free surface and keeping constant the water level by water refilling until the level obtained in the calibration-part during the whole certification-part of the procedure.

Secondly, this work deals with tests of pure-hydrogen gas meters, thus tests with pure-hydrogen will be presented. However, any other gas can be used due to the applicability of the model to any kind of gas. Indeed, the geometric volume obtained during calibration is constant and, thus, is the same also for each test performed after calibration, regardless of the nature of the gas. Additionally, also the calculation of the evaporated volumetric flow rate does not depend on the nature of the gas.

Finally, it is also possible to further validate the test bench by considering the alignment between different laboratories or test benches. Specifically, the standard ISO 17043:2010 defines a quantitative way to compare results from different test benches, by means of the dimensionless quantity  $E_n$  (-) that is calculated as follows:

$$E_n = \frac{|E_{WDM} - E|}{\sqrt{U_{WDM}^2 + U^2}} \quad (13)$$

where  $E_{WDM}$  (-) is the percentage error obtained with the new test bench,  $E$  (-) the percentage error calculated with an ISO 17025 certified labo-

ratory,  $U_{WDM}$  (-) the expanded uncertainty of the new test bench, and  $U$  (-) the expanded uncertainty assigned to ISO 17025 certified laboratory. A value of  $E_n$  lower than 1 suggests a good alignment between laboratories or test benches. However, more indexes for the determination of confidence levels should further support this analysis. The calculation of these indexes is outside the scope of this work and, thus, it is omitted.

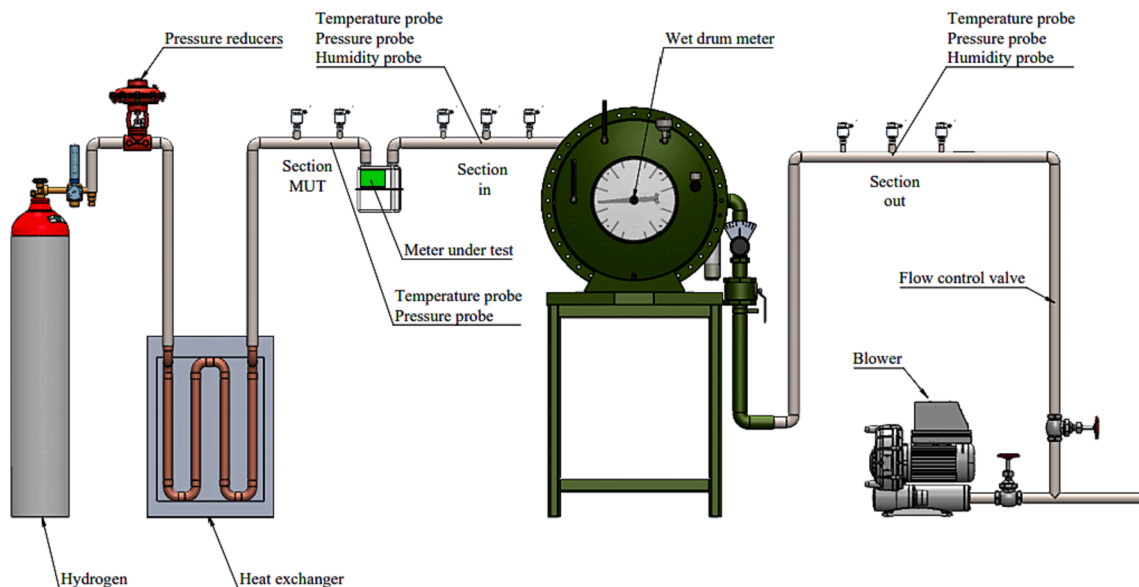
## 2.4. Test bench

During the calibration-part, the wet drum meter (WDM) is placed downstream of a bell prover for the calculation of the geometric volume and the tests are performed in air. Moreover, during the certification-part, the wet drum meter is placed downstream a meter under test and the tests are performed in hydrogen. Lastly, during the validation-part, both a bell prover and the wet drum meter are used to compare their volumetric flow rates with respect to the that of a travelling measurement standard, in this case, tests are performed both in air and helium.

In all the three parts of the procedure, temperature, pressure and relative humidity must be measured at the inlet and outlet sections of the wet drum meter to correct the volumetric flow rates as depicted in section 2.3. In this work, a diaphragm gas meter is employed as meter under test (MUT) during the certification-part and as travelling measurement standard during the validation-part. The characteristics of the employed wet drum meter and the MUT are reported in Table 1.

Hydrogen, air and helium are used in this work as test gases, while nitrogen as purge gas. Both hydrogen (purity  $\geq 99.9995$  %mol) and helium (purity  $\geq 99.998$  %mol) are extracted by certificated 200-bar cylinder tanks, while air is taken from the laboratory. The tests were performed in laboratory at conditions under control: temperature is kept at 20 °C within  $\pm 0,5$  °C, while pressure at atmospheric conditions of about 989 mbar with an uncertainty of 4 mbar and relative humidity 50%. Regulating valves are used to regulate the gas flow rate, that is then measured by the meters.

Fig. 1 shows a schematic of the test bench during the certification-part procedure, while Fig. 2 a picture of the same. The wet drum meter is located downstream of the diaphragm meter under test. Pure hydrogen volumes and volumetric flow rates are measured, while the relative error between the two instruments is calculated. The test bench comprises pressure transducers (measurement uncertainty of 5.8 Pa), temperature (measurement uncertainty of 0,06 °C) and humidity



**Fig. 1.** Scheme of the test bench used in the certification-part procedure to certify the hydrogen volumetric flow rate measurements of a diaphragm gas meter under test (MUT) G 4–10 against the calibrated reference wet drum meter (WDM).

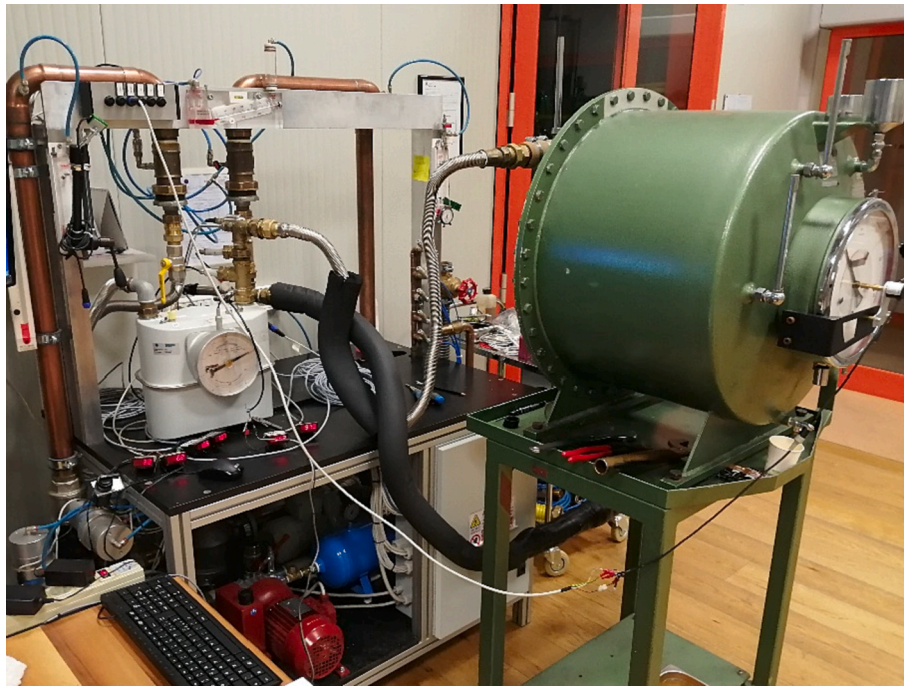


Fig. 2. Picture of the test bench used to compare the volumetric flow rate measurements of a diaphragm gas meter G 4–10 and the reference wet drum meter.

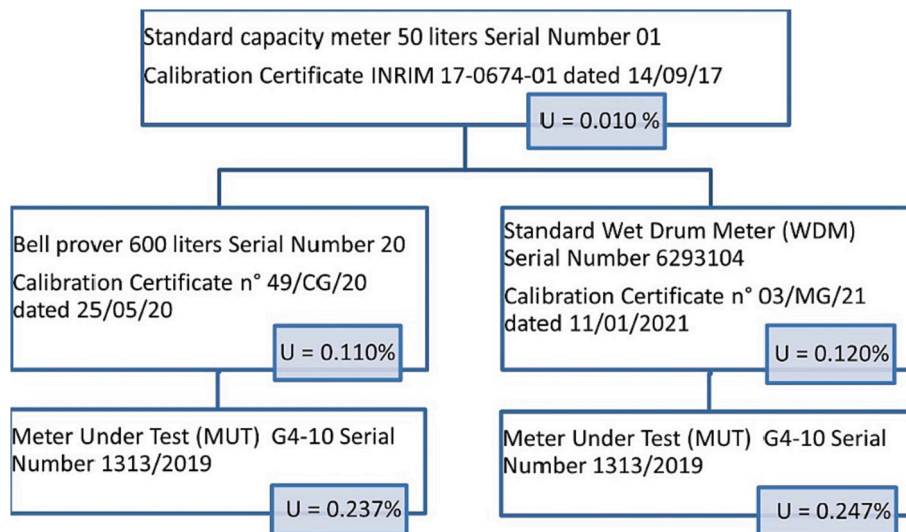


Fig. 3. Traceability scheme of the measurement procedure including a standard capacity meter, bell prover, wet drum meter and meter under test with their uncertainty contributions. The shown uncertainty values are derived from a real case and are meant to represent their order of magnitude.

sensors (measurement uncertainty of 2%), as well as regulating valves to control the flow rates. Pressure reducers and stabilizers, and a heat exchanger for temperature control are inserted in the test bench for the certification-part performed with hydrogen. Specifically, the stabilizer, also defined as gas regulator, allows to maintain a constant gas to air ratio even when the air pressure varies. For the calibration-part procedure, air volumes and volumetric flow rates are measured, while the geometric volume of the WDM is calculated. Pressure reducers and stabilizers, the heat exchanger and the MUT are not included in the test bench, and the hydrogen tank is substituted by a bell prover containing air. For the validation-part procedure, air or helium volumes and volumetric flow rates are measured, while the relative error of the travelling measurement standard with respect to the bell prover and WDM are calculated and compared. Pressure reducers and stabilizers, and the heat exchanger are not included in the test bench, and the hydrogen tank is

substituted by a bell prover containing air or helium. In this third part, the MUT is referred as travelling measurement standard. The exhausted hydrogen is vented outside of the laboratory by mixing hydrogen with air keeping the percentage of hydrogen below half of flammability level. As shown in Fig. 1, the temperatures and pressures are measured before the MUT, before the WDM, and after the WDM. The humidity is measured both before and after the WDM to account for water evaporation, while humidity measurement is not necessary in the MUT section. Finally, the employed PLC S7-1500 ET200S by Siemens can simultaneously activate several timers with a base frequency of 1024 kHz to measure the test time.

### 2.5. Measurement uncertainty calculation

First, the traceability of the test bench has to be defined to describe

**Table 2**

Expanded uncertainties and statistical distributions of the quantities directly measured in the test bench. Expanded uncertainties are characterized by a confidence interval of 95 % and derive from calibration certificates of the measurement instruments.

Measured quantity	Unit of measure	Expanded uncertainty ( $U$ )	Coverage factor	Statistical distribution
$t$	s	$10^{-6}$	$2\sqrt{3}$	rectangular
$n_{rev}$	rev/test	0.01	$2\sqrt{3}$	rectangular
$Q_{BLL}$	l/h	0.11	2	normal (Gaussian)
$T_{BLL}$	K	0.028	2	normal (Gaussian)
$P_{BLL}$	Pa	2.30	2	normal (Gaussian)
$T_{in}$	K	0.028	2	normal (Gaussian)
$P_{in}$	Pa	2.30	2	normal (Gaussian)
$RH_{in}$	%	1.10	2	normal (Gaussian)
$T_{out}$	K	0.028	2	normal (Gaussian)
$P_{out}$	Pa	2.30	2	normal (Gaussian)
$RH_{out}$	%	1.10	2	normal (Gaussian)

the uncertainty of measurement. The traceability scheme followed here is represented in Fig. 3. Specifically, the calibration of each instrument contributes to the uncertainty at each level of the traceability chain, from the standard capacity meter to the bell prover and WDM uncertainties that all contribute to the MUT uncertainty calculation. The metrological chain starts from a standard capacity meter, considered as primary reference and used for the calibration of the bell prover and wet drum meter. This allows to compare in a consistent way the MUT with both the bell prover and the WDM. The uncertainty of the standard capacity meter derives from INRIM [32] certificates and is equal to 0.010%. The uncertainty calculations are performed in accordance with a procedure that has been filed at a national accreditation body [33]. The uncertainty values shown in Fig. 3 are calculated for a real case, employing the relations described later in this section, and they are meant to represent the order of magnitude of the uncertainty of the proposed traceability scheme, but they can slightly change from

**Table 3**

Inlet and outlet pressure, temperature and relative humidity measurements, bell volumetric flow rates, wet drum meter revolutions and test duration calibration data for the wet drum meter geometric volume calculation. Tests are performed with air.

$Q'_{nom}$ (l/h)	$P'_{in}$ (kPa)	$P'_{out}$ (kPa)	$T'_{in}$ (K)	$T'_{out}$ (K)	$RH'_{in}$ (%)	$RH'_{out}$ (%)	$Q'_{BLL}$ (l/h)	$n'_{rev}$ (rev)	$t$ (s)	$V_{WDM}$ (l)
200	99.115	99.118	293.45	293.25	57	100	210.17	1	853.55	$50.33 \pm 0.030$
600	99.114	99.115	293.45	293.35	57	100	628.47	1	285.05	$50.27 \pm 0.022$
1200	99.114	99.115	293.45	293.30	57	100	1194.51	2	300.72	$50.39 \pm 0.021$
2400	99.112	99.109	293.40	293.35	58	100	2446.71	2	147.09	$50.49 \pm 0.020$
6000	99.110	99.106	293.30	293.20	58	100	6189.85	4	115.76	$50.26 \pm 0.020$

**Table 4**

Sources of uncertainties with weight coefficients and statistical distributions of the geometric volume calculation for the test at 6000 l/h for sake of example.

Measured quantity	Unit of measure	Quantity estimation	Standard uncertainty ( $u$ )	Weight coefficient	Coverage factor	Statistical distribution
$n'_{rev}$	rev/test	4	0.00002	29.22851	2	normal (Gaussian)
$Q'_{BLL}$	l/s	1.719	0.00866	0.00043	2	normal (Gaussian)
$T'_{BLL}$	K	293.3	0.00990	0.17136	2	normal (Gaussian)
$P'_{BLL}$	bar	0.991	0.00001	50.70690	2	normal (Gaussian)
$T'_{WDM}$	K	293.2	0.00001	50.70690	2	normal (Gaussian)
$P'_{WDM}$	bar	0.991	0.01400	0.17135	2	normal (Gaussian)
$Y'_{H2O,in}$	%	1.39	0.00013	50.96190	2	normal (Gaussian)
$Y'_{H2O,out}$	%	2.37	0.00013	51.47433	2	normal (Gaussian)

calibration to calibration.

The uncertainty of the measured data and the geometric volume must be investigated to calculate the uncertainty of the corrected gas volumetric flow rate of the wet drum meter. Measured data are retrieved from direct measurements performed by the laboratory instruments. Thus, their uncertainties are taken from the instrument calibration certificates provided by the manufacturers. Specifically, these are uncertainties of type B [34] with a normal statistical distribution. This is except for the encoder applied to the WDM which measures the number of revolutions for each test. Its uncertainty stems from a technical specification of the encoder and is considered with a rectangular statistical distribution. The expanded uncertainties,  $U$ , and the statistical distributions related to each measured quantity are summarized in Table 2.

Regarding the geometric volume uncertainty calculation, Eq. (6) is expanded to show the dependency of the geometric volume on all the main quantities. The final geometric volume expression is as follows:

$$V_{WDM} = \frac{t \cdot Q'_{BLL}}{n'_{rev}} \cdot \frac{P'_{BLL} T'_{WDM}}{T_{BLL} P'_{WDM}} \cdot \left( 1 + \frac{Y'_{H2O,out} - Y'_{H2O,in}}{1 - Y'_{H2O,out}} \right) \quad (14)$$

Thus, the calculation of the combined uncertainty is necessary, and is performed in accordance with the Guide to the Expression of Uncertainty in Measurement (GUM) considering all the terms in Eq.(14) except the time as a source of uncertainty. The combined standard uncertainty  $u_f$  of a function  $f(x_1, \dots, x_n)$  depends on the uncertainty of the  $x_i$  quantities involved in the model. Its value is calculated as follows:

$$u_f = \sqrt{\sum_{i=1}^n \left( \frac{\partial f(x_i)}{\partial x_i} u_{x_i} \right)^2} \quad (15)$$

where  $n$  is the number of the quantities,  $x_i$ . Then, the expanded uncertainties,  $U$ , is related to the standard uncertainty,  $u$ , as follows:

$$U = u \cdot K \quad (16)$$

where  $K (-)$  is the coverage factor related to the statistical distribution and the system degrees of freedom. In this study, the rectangular statistical distribution of the number of revolutions is normalized to a Gaussian distribution by dividing its expanded uncertainty by a  $2\sqrt{3}$  factor.

Applying Eqs. (15) and (16) to the geometric volume of Eq. (14), the expanded uncertainty of the geometric volume,  $U_{V_{WDM}}$  (l), is found. The



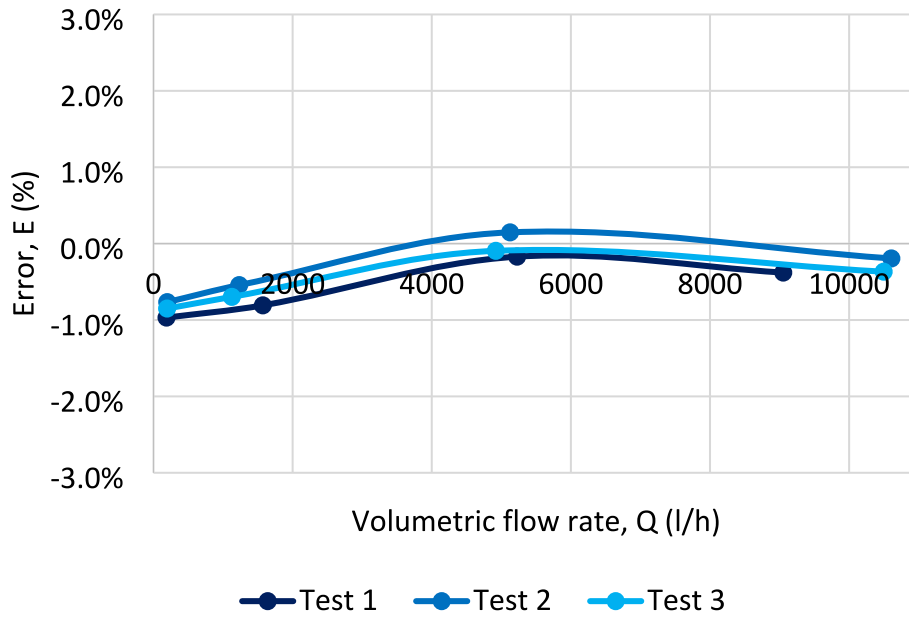


Fig. 4. Reading error curves of the Meter Under Test (MUT) with respect to the reference Wet Drum Meter (WDM) for three tests with hydrogen.

Table 5

Inlet and outlet pressure, temperature and relative humidity measurements, wet drum meter revolutions and test duration data for the calculation of the hydrogen corrected volumetric flow rate of the wet drum meter during Test 2, taken as example from the three tests.

$P_{in}''$ (kPa)	$P_{out}''$ (kPa)	$T_{in}''$ (K)	$T_{out}''$ (K)	$RH_{in}''$ (%)	$RH_{out}''$ (%)	$n_{rev}''$ (rev)	$t''$ (s)	$Q_{WDM}''^*$ (l/h)	$Q_{MUT}''^*$ (l/h)	$E$ (%)
100.91	100.84	292.85	292.35	6	100	1	907.61	$198.87 \pm 0.08$	197.35	-0.766%
100.87	100.80	293.45	292.35	6	100	2	300.92	$1238.7 \pm 0.5$	1231.96	-0.543%
100.73	100.66	293.75	292.35	6	100	2	90.63	$5118.7 \pm 2.2$	5126.25	0.147%
100.68	100.58	293.35	292.35	6	100	4	70.29	$10628 \pm 4$	10607.17	-0.194%

Table 6

Sources of uncertainties with weight coefficients and statistical distributions of the wet drum meter volumetric flow rate calculation for Test 2 at 6000 l/h for sake of example.

Measured quantity	Unit of measure	Quantity estimation	Standard uncertainty ( $u$ )	Weight coefficient	Coverage factor	Statistical distribution
$V_{WDM}$	l	50.347	0.00529	0.02824	2	normal (Gaussian)
$n_{rev}$	rev/test	2	0.00500	0.54402	2	normal (Gaussian)
$Y_{H2O.in}$	%	0.14	0.00013	1.42392	2	normal (Gaussian)
$Y_{H2O.out}$	%	2.2	0.00012	1.42392	2	normal (Gaussian)

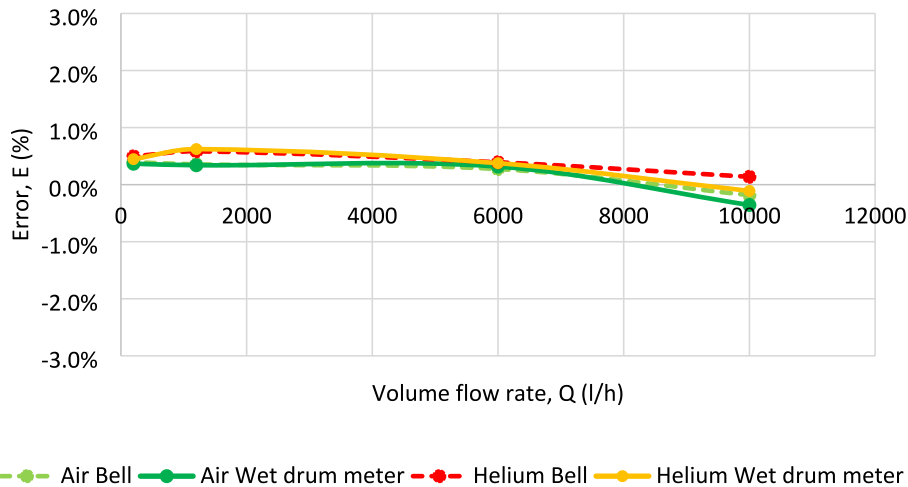


Fig. 5. Error curves in air and helium of the travelling measurement standard with respect to the bell prover and the wet drum meter.

**Table 7**

Values of the wet drum meter and bell prover errors and uncertainties, and the calculated quantity  $E_n$  at each tested flow rate in air.

$Q_{nom}$ (l/h)	$E_{WDM}$ (%)	$U_{WDM}$ (%)	$E_{BLL}$ (%)	$U_{BLL}$ (%)	$E_n$ (-)
200	0.37	0.32	0.39	0.30	0.06
1200	0.34	0.28	0.36	0.25	0.04
6000	0.32	0.27	0.27	0.25	0.12
10,000	-0.36	0.27	-0.18	0.25	0.49

**Table 8**

Values of the wet drum meter and bell prover errors and uncertainties, and the calculated quantity  $E_n$  at each tested flow rate in helium.

$Q_{nom}$ (l/h)	$E_{WDM}$ (%)	$U_{WDM}$ (%)	$E_{BLL}$ (%)	$U_{BLL}$ (%)	$E_n$ (-)
200	0.45	0.32	0.50	0.30	0.12
1200	0.62	0.28	0.58	0.25	0.11
6000	0.38	0.27	0.39	0.25	0.04
10,000	-0.11	0.27	0.14	0.25	0.70

determination of the geometric volume must be performed during calibration by way of tests at different flow rates. It is necessary to use air as calibration gas because required by standards, yet tests with other gases could be executed to verify the calculated geometric volume. Therefore, a mean value of the geometric volume,  $\overline{V_{WDM}}$  (l), must be computed, according to the total number of tests. Then, the corresponding expanded combined uncertainty is calculated through Eq. (15), which rearranged with Eq. (16) becomes as follows:

$$U_{\overline{V_{WDM}}} = K \cdot \sqrt{\sum_{i=1}^{n_{test}} \left( \frac{u_{V_{WDM,i}}}{n_{test}} \right)^2} \quad (17)$$

where  $n_{test}$  (-) is the total number of tests performed,  $U_{V_{WDM,i}}$  (l) the expanded uncertainty of each geometric volume resulted from each test. The coverage factor  $K$  is assumed equal to 2 due to the normalized statistical distribution of the quantities.

Finally, the geometric volume as well as the measurement uncertainties are used to calculate the test volumetric flow rate uncertainty with a similar procedure, i.e. applying Eqs. (15) and (16) to the corrected wet drum meter volumetric flow rate of Eq. (9).

### 3. Results

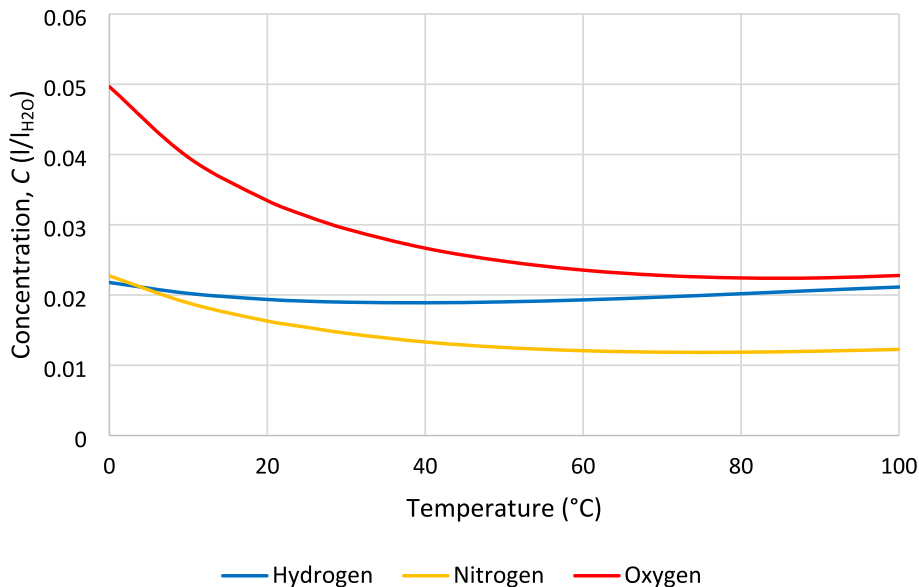
This section deals with the results of the present study. First, the values of the experimented flow rates are presented. Secondly, the wet drum meter geometric volume is calculated during the calibration-part using air. Then, the results of the certification-part using hydrogen are shown. Then, the procedure is validated performing tests with both air and helium. The values of the measurement uncertainty are shown for all the procedure parts.

#### 3.1. Experimented volumetric flow rates

The maximum volumetric flow rate for hydrogen,  $Q_{max}$ , is defined here by considering to triple the maximum rate for natural gas in domestic applications in order to transport the same quantity of energy given the fact that hydrogen energy content is roughly one third of natural gas on a volume basis at same temperature and pressure. Because the value of  $Q_{max}$  for natural gas is typically 6000 l/h in domestic applications, the value for hydrogen would be 18000–20000 l/h. Moreover, because the minimum volumetric flow rate,  $Q_{min}$ , is the 1/150 of the maximum as defined in European standards of natural gas meters, the value of  $Q_{min}$  for hydrogen would be 120–133 l/h. The adopted travelling meter operates at different volumetric flow rate ranges depending on the flowing fluid, e.g. between 60 and 10000 l/h for air and between 180 and 30000 l/h for hydrogen. Thus, the volumetric flow rates experimented in this work are in the mutual range from 200 to 10000 l/h. To cover a wider range of flow rates two travelling meters should have been used. However, this evaluation is outside the scope of this work, that aims at demonstrating the goodness of the procedure used.

#### 3.2. Calibration-part results

This section deals with the results of the wet drum meter calibration for the computation of its geometric volume performing the tests with air. During calibration, the wet drum meter inlet and outlet pressure,  $P_i$  (Pa), temperature,  $T_i$  (K), and relative humidity,  $RH_i$  (%), as well as the bell volumetric flow rate,  $Q_{BLL}$  (l/h), the number of revolutions per test,  $n_{rev}$  (rpm), and the test duration,  $t$  (s), are used to calculate the geometric volume,  $V_{WDM}$ , as described in Eq. (6) for different nominal flow rates,



**Fig. 6.** Saturation concentration of hydrogen, nitrogen and oxygen in water as a function of water temperature at 1 atm, redrawn after rearrangement based on data from Kolev [36].

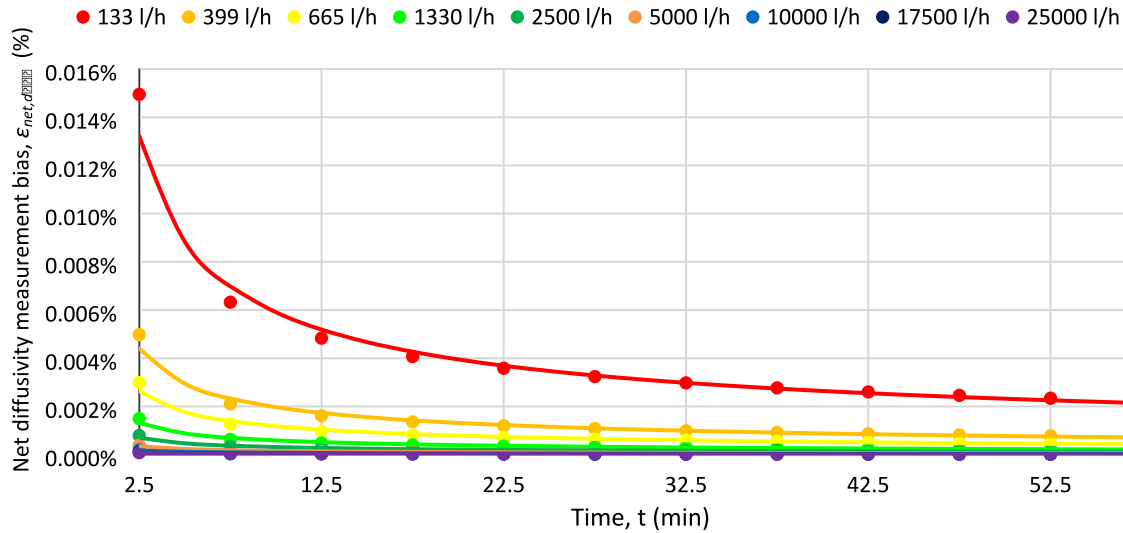


Fig. 7. Predicted biases of the volumetric flow rate measurements due to hydrogen diffusion during the first hour of test over a wide range of volumetric flow rates.

$Q_{nom}$  (l/h).

Table 3 shows these data retrieved from the experimental bench. The values of the geometric volumes of the different tests are used to calculate the mean geometric volume as well as the combined expanded uncertainty via Eq. (17), that turn out to be equal to  $(50.347 \pm 0.011)$  liters. Table 4 depicts the sources of uncertainty considered for the calculation of the geometric volume and anticipated in the previous section together with their weight coefficients and distribution. The main sources of uncertainty are the relative humidity and the temperature measurements.

### 3.3. Certification-part results

Once the calibration is completed, tests in hydrogen can be done using the wet drum meter as the reference meter. Water leveling of the wet drum is important to obtain better results as explained in section 2.3.1: water leveling should be equal to the one defined during calibration and refilling should be performed before each test. In this way, the actual geometric volume of the wet drum meter can be considered the same as that retrieved during calibration. The corrected wet drum meter volumetric flow rates,  $Q_{WDM}^{**}$ , are calculated by Eq. (9) starting from the geometric volume obtained via calibration. Also in this case, inlet and outlet values of pressure,  $P_i^*$  (Pa), temperature,  $T_i^*$  (K), and relative humidity,  $RH_i^*$  (%), as well as the number of revolutions per test,  $n_{rev}^{**}$  (rpm), and the test duration,  $t^*$  (s), are needed.

Fig. 4 shows the volume flow rate error curves of the MUT with respect to the WDM for three different tests. The trends of the curves are similar and characterized by a repeatability that is within 0.27% considering the maximum observed spread. The error curves are around zero, except at lower flow rates where errors are negative due to an expected behavior of diaphragm meter with low density gases such as hydrogen, as anticipated in section 2.3.3. For sake of numerical example, Table 5 shows the measured quantities as well as the calculated volumetric flow rates of hydrogen with their uncertainty obtained during Test 1. Table 6 depicts the sources of uncertainty considered in this work and anticipated in the previous section together with their weight coefficients and distribution for the wet drum meter volume flow rate calculation.

In order to quantify the effect of evaporation on the measurement,

the error that would be obtained from Eq.(1) comparing the MUT volumetric flow rate  $Q_{MUT}^*$  with the wet drum meter volumetric flow rate without the correction due to evaporation  $Q_{WDM}^{**}$  of Eq. (10) is calculated. As a numerical example, let's consider test 1 with a nominal flow rate of 200 l/h. In this case,  $Q_{MUT}^*$  is equal to 187.06 l/h and  $Q_{WDM}^{**}$  is calculated from Eq.(10) and the data in Table 4, and results to be equal to 192.81 l/h. The error is calculated using Eq. (1) and results to be as high as 3%. These errors are not acceptable in terms of metrological traceability, thus a detailed method to account for the evaporated water is necessary, as presented in this paper starting from the precise measurements of temperature, pressure and relative humidity at the inlet and at the outlet of the wet drum meter.

### 3.4. Validation-part results

In order to verify successful calibration, a travelling measurement standard is used to compare the behavior of the bell prover and the wet drum meter. Specifically, a comparison of the reading error curves of the travelling measurement standard with respect to the bell prover and wet drum meter is presented for both air and helium to validate the wet drum meter calibration and highlight the effect of the gas density on diaphragm gas meters. The corrected wet drum meter volumetric flow rates are calculated by Eq. (9) starting from the geometric volume obtained via calibration. Also this case requires inlet and outlet pressure,  $P_i^*$  (Pa), temperature,  $T_i^*$  (K), and relative humidity,  $RH_i^*$  (%), as well as the number of revolutions per test,  $n_{rev}^{**}$  (rpm), and the test duration,  $t^*$  (s).

Fig. 5 shows the error curves of the travelling measurement standard with respect of the bell prover in a certified laboratory according to ISO 17025 and using the wet drum meter with air and helium as flowing gases. The error curves of with respect to the bell prover and wet drum meter have maximum differences of 0.2% and 0.3% at the high flow rates for air and helium, respectively. On top of this, it is possible to notice that helium error curves are shifted upwards with respect to the air curves. This effect is caused by the lower density of helium with respect to air which influences the behaviour of the diaphragm: its membranes tend to deform less if the gas density is lower at the same flow rate, decreasing thus the measured cyclic volume and shifting the error to positive values. This phenomenon is more evident at the lowest

flow rates due to the further decrease of the filling velocity of the meter chambers. Thus, the authors suggest the possibility to adopt a smaller diaphragm and a larger one if using them as travelling measurement standards in executing the validation-part with a low density gas as helium.

These results confirm that the wet drum meter calibration procedure is performed successfully, validating the method. The error curves of the travelling measurement standard with respect to the bell prover and wet drum meter for a fixed flowing gas have maximum differences below 0.2–0.3%. Additionally, Table 7 and Table 8 show the values of  $E_n$  at each tested flow rate for air and helium, respectively. All the values are significantly lower than 1, thus a further investigation on the test bench compatibility is not necessary. This trend confirms the highly traceable alignment between the test benches, as showed also by Fig. 5.

#### 4. Conclusions

This study proposes a novel approach to the selection of the instrumentation and the definition of the procedure for the measurement of hydrogen volume as well as hydrogen volumetric flow rate in a legal framework. A new test bench equipped with a wet drum meter as the reference meter is designed and calibrated considering all sources of errors in the tests for type approval evaluation of new gas meters. A wet drum meter is selected as the reference meter due to its ability to measure both volume and volumetric flow rate of different flowing gases directly with the same accuracy. The procedure is divided into three parts: calibration, certification and validation, where:

- the calibration-part aims at calibrating the reference meter, that is the wet drum meter, via a bell prover in order to determine its geometric volume, which is the available volume occupied by the flowing gas in case the evaporation was not present and which is constant in all tests;
- the certification-part procedure aims at using the calibrated wet drum meter for the normal utilization of certifying any meter under test;
- the validation-part procedure aims at validating the calibration part and it is meant to be replicated only voluntarily at other laboratories.

The conclusions from this work are as follows.

- Water evaporation into hydrogen has an appreciable effect, introducing errors as high as 3% in the worst cases if not accurately addressed. These errors are not acceptable in terms of metrological

traceability, thus a detailed method to account for the evaporated water is presented here.

- The expanded uncertainty of the total metrological chain is in agreement with the international standards, *i.e.* OIML R137. It can be maintained below 0.25%.
- Hydrogen solubility and diffusivity into the water have no practical influence on the measurements. Indeed, the net measurement bias due to hydrogen diffusion in water with respect to air is up to 0.015%, as it will be demonstrated in Appendix A.
- The authors believe that the procedure is easily replicable in other laboratories and can be applied for different flowing gases, including pure gases and mixtures. In particular, it provides a new and reliable approach for hydrogen volume and flow rate measurement in a legal framework.
- The authors suggest using the instructions given in this work to perform type approval evaluation tests of new domestic gas meters compliantly with the current certification processes and can be useful for writing new international standards for gas meters including pure-hydrogen meters.

#### CRediT authorship contribution statement

**Camilla Nicol Bonacina:** Software, Formal analysis, Data curation, Writing – original draft, Visualization. **Paolo Tafani Alunno:** Conceptualization, Methodology, Validation, Investigation, Resources. **Massimo Mastromatteo:** Conceptualization, Methodology, Formal analysis, Writing – original draft. **Gianluca Valenti:** Conceptualization, Methodology, Writing – review & editing, Supervision. **Gilberto Serafini:** Conceptualization, Methodology, Validation, Investigation, Resources. **Claudio Imboccioli:** Conceptualization. **Matteo Russo:** Software, Formal analysis, Data curation. **Caterina Soranno:** Software, Formal analysis, Data curation. **Lorenzo Mori:** Validation, Investigation, Resources. **Filippo Maria Lombardi:** Data curation, Writing – review & editing.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Acknowledgement

None.

#### Appendix A

A portion of the gas flowing through the WDM diffuses into the water of the WDM itself. This section deals with gas diffusion in water for determining the volumetric flow rate measurement bias between the WDM and the MUT. Both hydrogen and air diffusion are studied. Indeed, hydrogen is the fluid under test during the certification-part, while air is the gas used to calibrate the wet drum meter in the calibration-part. Thus, the main quantity of interest of this study is net volumetric flow rate measurement bias of hydrogen with respect to air, being the effect of air diffusion in water already considered by the calibration procedure. First, the data of solubility and diffusivity of hydrogen and air into water from literature are presented. Then, the mathematical and numerical models, as well as their implementation, are analyzed. Lastly, their results are discussed, proving that the diffusivity bias can be neglected. The analysis omits helium for brevity, which would lead anyhow to the same conclusion.

##### Solubility and diffusivity data

The solubility and diffusivity coefficients of hydrogen and air in water must be investigated to describe quantitatively the effect of hydrogen and air diffusion in the water of the wet drum meter.

Regarding the hydrogen solubility, the studies of Young [35] and of other authors [36,37,38] are compared to analyze the experimental data and empiric relations of the solubility of hydrogen in water. In Young's work [35], the best fitting equation for 75 data points from other experimental works is obtained, and the hydrogen mole fraction solubilities at a gas partial pressure of 101325 Pa are computed, giving a standard deviation of 0.52% at the middle of the temperature range considered. Kolev [36] correlates hydrogen solubility data for temperatures up to 100 °C and pressure of 1 bar with an analytical expression with a mean error of 0.46%. Crozier and Yamamoto [37] determine Bunsen solubility coefficients of hydrogen in distilled water at atmospheric partial pressure of the gas as function of the measurement temperature with an estimated accuracy of 0.5%. Theodore

*et al.* [38] report Henry's constants at moderate pressure conditions, highlighting that the pressure dependence can be neglected for pressures up to 5 bar. All these literature studies show results in close agreement. Moreover, Kolev [13] offers a comparison among hydrogen, nitrogen, and oxygen solubility data, useful to relate the behaviour of nitrogen and oxygen with respect to hydrogen. This aspect jointly with the agreement among hydrogen solubility data of the above-mentioned authors in the temperature range of interest leads to selecting the solubility values from this work. The raw data of Kolev [36] are rearranged to obtain concentrations as liter of hydrogen ( $\text{H}_2$ ) per liter of water ( $\text{H}_2\text{O}$ ) at actual temperature and pressure conditions. Consequently, Fig. 6 shows the saturation concentration of hydrogen, nitrogen and oxygen as function of water temperature at 1 atm as obtained after the rearrangement. A hydrogen solubility of  $0.0194 \text{ l}_{\text{H}_2}/\text{l}_{\text{H}_2\text{O}}$  is found at  $20^\circ\text{C}$  and 1 bar.

Regarding the air solubility, the work of Battino [39] is investigated. Specifically, the Bunsen solubility coefficients of dried air in distilled water at 101325 Pa and 293.15 K with an estimated error of 1% are reported. Similarly to hydrogen, the author's data are rearranged to obtain the air concentration in water expressed as liter of air ( $\text{l}_{\text{air}}$ ) per liter of water ( $\text{l}_{\text{H}_2\text{O}}$ ). As a result, an air solubility of  $0.02 \text{ l}_{\text{air}}/\text{l}_{\text{H}_2\text{O}}$  at  $20^\circ\text{C}$  and 1 bar is found. Hence, the difference between hydrogen and air solubility is in the order of  $10^{-4}$ .

Regarding the diffusivity, the results of the works of Houghton *et al.* [40], Vivian and King [41], Wise and Houghton [42], Ferrell and Himmelblau [43], as well as Mazarei and Sandall [44] are compared. The cited authors investigate the diffusion phenomenon of hydrogen in water to reach equilibrium conditions of the flow. Specifically, the most recent study [44] employs a wetted wall column technique for measuring and calculating some parameters from which the hydrogen diffusion coefficient can be computed. Experiments with carbon dioxide gas, with a well-known diffusion coefficient, are also carried out to validate this method. They found an average hydrogen diffusion coefficient of  $4.8 \cdot 10^{-5} \text{ cm}^2/\text{s}$  at  $25^\circ\text{C}$ . This result is in accordance with the results of the previous works of Vivian and King [41], Ferrell and Himmelblau [43] and Houghton *et al.* [40]. Among these literature references, Houghton *et al.* [40] provide also the air diffusion coefficient in water at  $19.6^\circ\text{C}$ , which is equal to  $2.3 \cdot 10^{-5} \text{ cm}^2/\text{s}$ .

### Diffusivity model and solution

During the tests, hydrogen flows through the wet drum meter, that is partially filled with water. Thus, a portion of hydrogen diffuses from the water free surface into water itself. The rate of diffusion changes from the beginning of the tests, where no hydrogen is present in water, to the end, where saturation is reached. Both diffusion and advection affect hydrogen concentration in water due to the presence of a hydrogen flow at the water free surface. The scope of the following model is to estimate the order of magnitude of diffusion over the whole test duration to estimate the biases both at the beginning and at the end of the tests, and to find out when equilibrium conditions are reached.

A one-dimension (1D) approach is considered to evaluate the order of magnitude of the measurement bias obtained due to hydrogen diffusion in water. Hence, the 1D advection–diffusion equation is selected to describe the transport of gas in water as function of time and position [45]:

$$\frac{\partial C}{\partial t} + v \frac{\partial C}{\partial x} = D \frac{\partial^2 C}{\partial x^2} \quad (18)$$

where  $C$  ( $\text{l}_{\text{gas}}/\text{l}_{\text{H}_2\text{O}}$ ) is the gas concentration and  $v$  (m/s) the gas flow velocity. The gas velocity is assumed constant and one-dimensional for all tests. Moreover, it is calculated for each test as a function of the volumetric flow rate and the orifice area through which hydrogen enters the WDM. The depth of the WDM is computed from the geometrical drawing of the instrument. The gas diffusion coefficient at the laboratory operating temperature,  $D$  ( $\text{m}^2/\text{s}$ ), is assumed also to be constant and calculated from the literature reference value of diffusion coefficient at  $25^\circ\text{C}$  by the Stokes-Einstein theory as follows:

$$\frac{D\mu}{T} = \frac{k}{6\pi r} \quad (19)$$

where  $\mu$  ( $\text{Pa} \cdot \text{s}$ ) is the solvent viscosity,  $T$  (K) is the solution temperature,  $k$  (J/K) the Boltzmann constant, and  $r$  (m) the hydrodynamic radius of the diffusing particles [44,41,46]. This relation indicates that the diffusion coefficient decreases with decreasing temperature for small concentrations of the solute in the solvent [38].

The implicit 1D Crank-Nicolson method is chosen to solve the time-dependent advection–diffusion equation [47]. The Crank-Nicolson method is a combination of the backward and forward Euler's methods and guarantees numerical stability. Mathematically:

$$\frac{C_j^{n+1} - C_j^n}{\Delta t} + u \left[ \frac{1}{2} \left( \frac{C_{j+1}^{n+1} - C_{j-1}^{n+1}}{2\Delta x} + \frac{C_{j+1}^n - C_{j-1}^n}{2\Delta x} \right) \right] = D \left[ \frac{1}{2} \left( \frac{C_{j+1}^{n+1} - 2C_j^{n+1} + C_{j-1}^{n+1}}{\Delta x^2} + \frac{C_{j+1}^n - 2C_j^n + C_{j-1}^n}{\Delta x^2} \right) \right] \quad (20)$$

where  $j$  is the spatial index ranging between 1 and  $J$ ,  $n$  the time index ranging between 1 and  $N$ , while  $\Delta t$  and  $\Delta x$  are the temporal and spatial steps, respectively. Space is discretized along the depth of water in the wet drum in  $J$  control volumes.

Moreover, introducing the following constants:

$$\begin{cases} \lambda \stackrel{\text{def}}{=} \frac{D\Delta t}{2\Delta x^2} \\ \alpha \stackrel{\text{def}}{=} \frac{u\Delta t}{4\Delta x} \end{cases} \quad (21)$$

Eq. (20) is simplified as follows:

$$-(\lambda + \alpha)C_{j-1}^{n+1} + (1 + 2\lambda)C_j^{n+1} - (\lambda - \alpha)C_{j+1}^{n+1} = (\lambda + \alpha)C_{j-1}^n + (1 - 2\lambda)C_j^n + (\lambda - \alpha)C_{j+1}^n \quad (22)$$

The following boundary and initial conditions are set to solve the linear system of equations:

$$\begin{cases} C(j=1, n) = C_{\text{sat}} \\ C_{j-1}^n = C_{j+1}^n \\ C(j \neq 1, n=0) = 0 \end{cases} \quad (23)$$

where  $C_{sat}$  ( $l_{H2}/l_{H2O}$ ) is the gas saturation concentration and the spatial index  $j$  is set equal to 1 at the water free surface. Finally, the expression of linear system of equations in a matrix form leads to the final solution as follows:

$$[C^{n+1}] = [AA^{-1}][BB][C^n] + [d] \quad (24)$$

where  $[C^{n+1}]$  and  $[C^n]$  are vectors of  $J+1$  dimension containing the gas concentration values at the time-steps  $n+1$  and  $n$ , respectively;  $[AA^{-1}]$  is the inverse of the  $[AA]$  tridiagonal matrix [48] with the coefficients of the linear system corresponding time-step  $n+1$ ;  $[BB]$  is the tridiagonal matrix with the coefficients of the linear system corresponding to the time-step  $n$ ;  $[d]$  is the vector of  $J+1$  dimension containing the initial and boundary conditions. The Crank-Nicolson model is implemented in a MATLAB code and used to simulate the diffusivity behavior of both hydrogen and air in water under the conditions of the present investigation.

Finally, the Crank-Nicolson model is validated comparing the obtained results with those of the exact analytical solution, that considers diffusion only [45], because advection effects seem negligible over the whole test cycle. Specifically, in the range between 25 and 500 days, 0 and 37.5 cm depth, the maximum errors obtained for hydrogen and air diffusion are equal to 0.2% and 1.1%, respectively, at 37.5 cm depth after 25 days of hypothetical operation. Thus, the implementation is validated, and its results can be considered acceptable in the whole range of measurements.

The simulations are executed until water is completely saturated with the gas. The output of the model consists in the prediction of gas concentration in water as a function of time and depth of the wet drum meter. Moreover, the volumetric flow rate of gas diffused in water,  $Q_{diff}$  (l/h), is computed for different time intervals as follows:

$$Q_{diff} = \frac{(\bar{C}^{n+1} - \bar{C}^n)V}{t^{n+1} - t^n} \quad (25)$$

where  $V$  (l) is the volume of water contained in the wet drum, while  $\bar{C}$  ( $l_{H2}/l_{H2O}$ ) is the mean gas concentration in water calculated with the trapezoidal integration function in MATLAB. Then, the diffusivity measurement bias,  $\varepsilon_{diff}$  (-), is calculated to depict the effect of diffusivity on the volumetric flow rate measurement as follows:

$$\varepsilon_{diff} = \frac{Q_{diff}}{Q_{test}} \quad (26)$$

where  $Q_{test}$  (l/h) is the test volumetric flow rate, that is in the range between the minimum and maximum volumetric flow rates,  $Q_{min}$  (l/h) and  $Q_{max}$  (l/h). Finally, the net volumetric flow rate measurement bias of hydrogen with respect to air is calculated as follows:

$$\varepsilon_{diff.net} = \frac{Q_{diff.H2} - Q_{diff.air}}{Q_{test}} = \varepsilon_{diff.H2} - \varepsilon_{diff.air} \quad (27)$$

### Diffusivity results and considerations

Hydrogen and air volumetric flow rates simulated here span from 133 to 25000 l/h, resulting even in a larger range than that experimented in section 3.1. Moreover, the WDM water depth and volume are measured experimentally and are found to be equal to 38 cm and 108 l, respectively. The gas inlet orifice is placed far above the liquid level, thus the advection term  $\alpha$  is considered null. Finally, the diffusion coefficient corrected at 20 °C is calculated by Equation (19), and is found to be equal to  $4.2 \cdot 10^{-5}$  cm<sup>2</sup>/s and  $2.3 \cdot 10^{-5}$  cm<sup>2</sup>/s for hydrogen and air, respectively.

Fig. 7 depicts the results from the diffusivity model. The whole depth of the wet drum meter is considered, and the results are reported each five minutes for 1 h of test. The net measurement bias,  $\varepsilon_{diff.net}$ , due to hydrogen diffusion in water with respect to air is up to 0.015% only in the most stringent case of minimum volumetric flow rate after 5 min. In any other cases, the bias is lower than 0.005% and tends to almost null values for higher volumetric flow rates. This result confirms that hydrogen diffusion in the wet drum does not represent a problem in terms of metrological traceability even when executing tests with new and clean water. Indeed, the diffusivity bias is at least one order of magnitude lower than measurement errors illustrated in Section 3. Finally, the simplification of the diffusivity model by way of the Crank-Nicolson method with a 1D approach is acceptable because of the highly moderate measurement bias obtained. Similar considerations could be obtained also for helium diffusivity in water with respect to air because hydrogen and helium are characterized by diffusivity coefficients of the same order of magnitude, as measured by Verhallen *et al.* [49].

### References

- [1] Paris Agreement Under the United Nations Framework Convention on Climate Change, Paris, 2015.
- [2] International Energy Agency, "The Future of Hydrogen," 2019.
- [3] G20 Tokyo Statement 2018, Tokyo, 2018.
- [4] R. Judd, D. Pinchbeck, Hydrogen admixture to the natural, in: *Compendium of Hydrogen Energy*, Woodhead Publishing, 2016, pp. 165-192.
- [5] J. Jaworski, P. Kulaga, T. Blacharski, Study of the Effect of Addition of Hydrogen to Natural Gas on Diaphragm Gas Meters, *Energies* 13 (11) (2020) 3006.
- [6] J. Jaworski, A. Dudek, Study of the Effects of Changes in Gas Composition as Well as Ambient and Gas Temperature on Errors of Indications of Thermal Gas Meters, *Energies* 13 (20) (2020) 5428.
- [7] M. Dell'Isola, G. Ficco, L. Moretti, J. Jaworski, P. Kulaga, E. Kukulska-Zajac, Impact of Hydrogen Injection on Natural Gas Measurement, *Energies* 14 (24) (2021) 8461.
- [8] J. Dehaeseleer, The Effects of Injecting Hydrogen (Renewable Gases), in: *EASEE-Gas GMOM*, Budapest, 2018.
- [9] G. Müller-Syring, M. Henel, W. Köppel, H. Mlaker, M. Sterner, T. Höcher, Entwicklung von Modularen Konzepten Zur Erzeugung, Speicherung und Einspeisung von Wasserstoff und Methan in Erdgasnetz, in *DVGW*, Bonn, 2013.
- [10] A.A. Stetsenko, S.D. Nedzelsky, V.A. Naumenko, The Effect of Hydrogen on the Physical Properties of Natural Gas and the Metrological Characteristics of Its Metering Systems, *Metrology and instruments* 6 (2020) 45–50.
- [11] "Hy4Heat," [Online]. Available: <https://www.hy4heat.info/>. [Accessed 09 November 2020].
- [12] "NewGasMet," Empir Euramet, [Online]. Available: <https://newgasmet.eu/>.
- [13] EMPIR, "Metrology infrastructure for high-pressure gas and liquified hydrogen flows," [Online]. Available: <https://www.methyinfra.ptb.de/>. [Accessed 25 03 2022].
- [14] EMPIR, "MetroHyVe 2 - Metrology for Hydrogen Vehicles 2," [Online]. Available: <https://www.sintef.no/projectweb>. [Accessed 25 03 2022].
- [15] J. van der Grinten, B. Mickan, H. Riezebos, D. van Putten, "Gas flow traceability for non-conventional and renewable gases," in *North Sea Flow Measurement Workshop*, 2021.
- [16] M. MacDonald, D. Anderson, T. Isaac, S. Mahmood, "Accuracy testing of domestic gas flow meters with 20% hydrogen blend," in *North Sea Flow Measurement Workshop*, 2021.
- [17] H. Riezebos, M. al Saleem, D. van Putten, R. ten Cate, JIP renewable gases; results on performance of turbine, in: *North Sea Flow Measurement Workshop*, 2021.
- [18] TUV SUD, "Hydrogen Flow Calibration Facility for Domestic Gas Meters," TUV SUD, 08 2020. [Online]. Available: <https://www.tuvsud.com/en-gb/-/media/regions/uk/pdf-files/industries/chemical-and-process/hydrogen-domestic-gas>

- meter-facility—marc-macdonald—tv-sd-nel.pdf?la=en-gb&hash=261DE9C9E2F446B82019CBCD8A3F01DC. [Accessed 24 02 2022].
- [19] International Organization of Legal Metrology, *OIML R 137-1 & 2*, 2012.
- [20] International Organization of Legal Metrology, *OIML R 140*, 2007.
- [21] European Commission, *Blue Guide*, Official Journal of the European Union, 2016.
- [22] The European Parliament and Council of the European Union, *Regulation (EC) 765/2008*, 2008.
- [23] The European Parliament and Council of the European Union, *Measuring Instruments Directive (MID) 2014/32/EU*, 2014.
- [24] F. Cascetta, M. Comazzi, M. Musto, G. Rotondo, An experimental intercomparison of gas meter calibrations, *Measurement* 45 (8) (2012) 1951–1959.
- [25] R.C. Baker, *Flow measurement handbook, SEZIONE IDROGENO (2000)*.
- [26] L.V. King, On the Convection of Heat from Small Cylinders in a Stream of Fluid: Determination of the Convection Constants of Small Platinum Wires, with Applications to Hot-Wire Anemometry, in: *Proceedings of the Royal Society of London - Series A*, 1914.
- [27] Ritter, “Drum-type gas meters,” [Online]. Available: <https://www.ritter.de/en/downloads/drum-type-gas-meters>. [Accessed 15 10 2021].
- [28] K. Park, H. Choi, Y. Oh, D. Lee, Performance Enhancement of Wet Gas Flow Meter, in: ASME 2002 Joint U.S.-European Fluids Engineering Division Conference, 2002.
- [29] J.N. Smith, H.C. Van Ness, M.M. Abbott, M.T. Swihart, *Introduction to chemical engineering thermodynamics*, McGraw Hill's (2017).
- [30] IAPWS, “The International Association for the Properties of Water and Steam,” [Online]. Available: <http://iapws.org/>. [Accessed 26 03 2022].
- [31] M.J. Moran, H.N. Shapiro, D.D. Boettner, M. Bailey, *Principles of Engineering Thermodynamics*, John Wiley & Sons, 2015.
- [32] INRIM, “Istituto Nazionale di Ricerca Metrologica,” [Online]. Available: <https://www.inrim.it/>. [Accessed 03 26 2022].
- [33] Accredia, [Online]. Available: <https://services.accredia.it>. [Accessed 06 07 2022].
- [34] E.O. Doebelin, *Strumenti e metodi di misura*, McGraw-Hill Companies, Milano, 2008.
- [35] C.L. Young, Hydrogen and Deuterium, in *Solubility Data Series: Vol. 5/6*, 1981.
- [36] N.I. Kolev, Solubility of O<sub>2</sub>, N<sub>2</sub>, H<sub>2</sub> and CO<sub>2</sub> in water, in: *Multiphase Flow Dynamics 4*, Springer, 2011, pp. 209–231.
- [37] T.E. Crozier, S. Yamamoto, Solubility of Hydrogen in Water, Seawater, and NaCl Solutions, *J. Chem. Eng. Data* 19 (1974) 242–244.
- [38] L.B. Theodore, S.L. Adrienne, P.I. Frank, P.D. David, *Fundamentals of Heat and Mass Transfer*, John Wiley & Sons, 2011.
- [39] R. Battino, Nitrogen and Air, *IUPAC Solubility Data Series* (1984).
- [40] G. Houghton, P.D. Ritchie, J.A. Thomson, The rate of solution of small stationary bubbles and the diffusion coefficients of gases in liquids, *Chem. Eng. Sci.* 17 (1962) 221–227.
- [41] J.E. Vivian, C.J. King, Diffusivities of Slightly Soluble Gases in Water, *AIChE J.* 10 (1964) 220–221.
- [42] D.L. Wise, G. Houghton, The diffusion coefficients of ten slightly soluble gases in water at 10–60 °C, *Chem. Eng. Sci.* 21 (1966) 999–1010.
- [43] R.T. Ferrell, D.M. Himmelblau, Diffusion coefficients of hydrogen and helium in water, *AIChE J.* 13 (1967) 702–708.
- [44] A.F. Mazarei, O.C. Sandall, Diffusion Coefficients for Helium, Hydrogen and Carbon Dioxide in Water at 25 °C, *AIChE J.* 26 (1980) 154–157.
- [45] F.P. Incropera, D.P. Dewitt, T. Bergman, A.S. Lavine, *Fundamentals of Heat and Mass Transfer*, John Wiley & Sons, U.S.A., 2006.
- [46] M. Cappelazzo, C.A. Capellari, S.H. Pezzin, L.A.F. Coelho, Stokes-Einstein relation for pure simple fluids, *J. Chem. Phys.*, 126(22), 2007.
- [47] M. Schafer, *Computational engineering. Introduction to numerical methods*, Springer Berlin Heidelberg New York, 2006.
- [48] W.H. Press, S.A. Teukolsky, W.T. Vetterling, B.P. Flannery, *Numerical Recipes in C, The Art of Scientific Computing*, Cambridge University Press, 1988.
- [49] P. Verhallen, L. Oomen, A.V. Elsen, A. Krugger, The diffusion coefficients of helium, hydrogen, oxygen and nitrogen in water determined from the permeability of a stagnant liquid layer in the quasi-steady state, *Chem. Eng. Sci.* 39 (11) (1984) 1535–1541.