The EMPIR initiative is co-funded by the European Union's Horizon 2020 research and innovation programme and the EMPIR Participating States

EMPIR Grant Agreement number	16ENG09
Project short name	LNG III
Project duration	2017-06-01 to 2020-06-30
Deliverable reference	*D5*
Deliverable title	*Recommendations for an SI-traceable density calibration method and cost effective (in-line) density sensors*
Involved partners (Lead partner in bold)	***INRiM**, NEL, VSL, JV*
Due date of the deliverable	*M35*
Actual submission date of the deliverable	M36

Metrological support for LNG and LBG as transport fuel

Authors: P. A. Giuliano Albo (INRiM), A. Kenbar (NEL), M. Schakel (VSL) and K. Hadidi (JV)

Recommendations for an SI-traceable density calibration method and cost effective (in-line) density sensors

P. A. Giuliano Albo (INRiM), A. Kenbar (NEL), K. Hadidi (JV) and M. Schakel (VSL)

1. Introduction

This document exploits the knowledge and experience obtained during the development of the EMPIR 16ENG09 LNG3 project to recommend an SI-traceable density calibration method using in-line sensors capable of measuring the density of flowing LNG. The attempt is to achieve the goals that have the potential to speed up the innovation in the field of LNG custody transfer measurements and controls. The document starts with an overview of the current state-of-the art for LNG density measurement methods.

Limited to the frame of density measurements, just a few years ago, it was not possible to obtain accurate and traceable results at cryogenic temperatures in LNG installations. Today a new hydrostatic weighing densimeter has been realized at RUB [1] so that it is possible to characterize reference materials and certified mixtures both to improve the equation of state of LNG and to calibrate secondary standards that can be used in LNG installations. At INRiM an innovative ultrasonic densimeter was developed that can be calibrated in the laboratory with a cryogenic reference fluid at rest and then it can be used as a secondary standard for SI-traceable LNG density measurements to calibrate density sensors used in flow metering facilities. The world's first metrological cryogenic facility developed by VSL can now operate with liquid nitrogen [2,3] and soon with LNG. This facility will provide the opportunity to characterize the ultrasonic densimeter indicated above at different flow rates, following the same approach adopted at NEL where density measurements using Coriolis flow meters have been carried out [4]. This ultrasonic sensor can be calibrated in the same way at ambient and cryogenic conditions, for the first time, for both flow rate and density.

The availability of innovative facilities allows also to obtain insight into behaviors of on-line sensors that had been difficult to assess before. This will provide the opportunity to take advantages of on-line measurement capabilities that have never been exploited before.

As part of the ongoing work, the simultaneous cryogenic speed of sound measurement device was completely validated and the results are reported by Cavuoto el al., 2019 [5].

The following sections describe the (in-line) technologies available for measurement of LNG density and the best practice to be adopted for achieving reliable and traceable measurements that meet current industry requirements. The focus will be on Coriolis and ultrasonic density measurements technologies.

2. State-of-the LNG density measurement methods[1]

In current industry practice where the LNG density is determined from the composition measurement, important uncertainty sources are the representative nature of the sample both in space and time, the choice of the correct equation of state, and the accuracy of temperature and composition measurements. The LNG industry would benefit from a reliable, and industry accepted, online and direct LNG density measurement, in which temperature and composition measurements are not required thus eliminating their contribution to the measurement uncertainty of the density.

A review of density calculation methods used in LNG custody transfer applications has been reported in [7]. Comparison of thirteen density calculation methods using open literature data for measured density showed that the method adopted by the LNG industry [8, 9], namely revised Klosek and McKinley method (RKM) appeared to give best accuracy for LNG density calculation. The uncertainty of the RKM method was reported to be 0.1%. This uncertainty estimate is only valid for calculations around atmospheric pressure. At higher pressures, experimental data have demonstrated that the RKM method is less accurate [10].

This method may also be applied for density calculations in small scale operations, however, some of these operations may be performed at pressures higher than the atmospheric pressure, corresponding to the saturation pressure for typical LNG mixtures at temperatures of about 110 K (-163 °C). As the RKM does not take pressure into account, it was necessary to adapt this method; the result is known as the Enhanced Revised Klosek and McKinley (ERKM) Method which has been developed by Ruhr University of Bochum (RUB) [10, 11]. This method can be used up to 100 bar.

The pressure-dependence of the ERKM method was obtained by fitting an additional term to densities of various typical LNG-mixtures calculated with the GERG-2008 equation of state [12]. Afterwards, the performance of the ERKM method was verified by means of highly accurate experimental data sets [10,11], using RUB's newly developed densimeter [13]. The uncertainty of ERKM method is the same as of the RKM, namely 0.1% at temperatures between 100 K (-173 °C) and 115 K (-158 °C). This uncertainty rises to 0.15% for temperatures from 115 K (-158 °C) to 135 K (-138 °C). The limitations of the RKM method in regard of the LNG components do also apply to this method.

Although, the primary densimeter developed by RUB provides highly accurate SI-traceable measurements of LNG density with estimated uncertainty of 0.02% [13], it cannot be used in the field to measure the LNG density directly as it has been designed as a laboratory device suitable for density measurement of LNG samples or pre-prepared LNG mixtures. It can also be used as a calibration reference or as a transfer standard for devices measuring LNG density in the field.

The methods described above for LNG density estimation is a summary of currently recommended methods by the LNG industry. However, interest in development of accurate and cost-effective means for measuring LNG density directly and without the need for sampling is growing [14].

A different measurement device for analysing LNG composition based on direct analysis in the LNG

[1] This section is taken from [6]

transfer line(s) hence eliminating the need of an LNG sampling, vaporisation and gas analysis equipment, is being tested in a few pilot applications and known as the Raman spectroscopy method. Raman spectroscopy is an analytical technique that uses monochromatic light to excite and identify the vibrational modes of molecules. Each mode of each molecule generates a shift in the frequency of the scattered light. By analysing the frequency and intensity of the scattered light, the sample's composition may be determined. The scattering interaction is so short-lived that the measurement is independent of the flow rate of the sample [8]. When validated for LNG composition measurement, this method can potentially result in determination of LNG density with better uncertainty than the traditional method which requires a representative sampling technique.

One of the currently available devices for direct measurement of fluid density on-line is by using a Coriolis flow meter. Coriolis meters have been shown to measure density of fluids such as water and oils with good accuracy due to availability of reference calibration facilities. However, when used under cryogenic conditions, the current-industry practice is to calibrate the meter with water for mass flow rate and density and then transfer this calibration to cryogenic conditions using appropriate corrections. References [2, 15] presented Coriolis flow meter calibrations at ambient and cryogenic conditions, however, no direct SI-traceable measurement of density under cryogenic conditions was performed as part of this work.

2. Coriolis sensors used for density measurements

Coriolis mass flow meters are supporting industrial activities in different fields, from food and beverage to oil and gas passing through chemical and pharmaceuticals, thanks to their capability to measure the true mass flow directly. Moreover, their stability and accuracy suggest that, with some efforts, these sensors would be able to extend their capabilities to measure the density of LNG at cryogenic temperature. To support further investigations, accurate calibration procedures and suitable simplified models, based on the experience maturated during the development of the 16ENG09-LNG3 project, are proposed.

2.1 Fluid density working equation

As a first raw approximation, the free oscillation frequencies of a vibrating tube filled by a fluid, can be described as a mass-spring discrete oscillatory system. In the case of small oscillations out of an equilibrium point, the expression for the frequency *f* has the form:

$$f = \frac{1}{2\pi}\sqrt{\frac{K}{M}} \tag{1}$$

where *M* is the mass and *K* is the elastic constant of the spring. As a little improvement of the model, it is possible to express the mass $M = \rho V + m$ in terms of the fluid density ρ, its volume *V* and the mass of the tube *m.* Rewriting equation (1), the following expression is obtained for the density of the fluid:

$$\rho = \frac{K_n}{4\pi^2 V {f_n}^2} - \frac{m}{V}. \tag{2}$$

In the expression (2), an index *n* has been added to *K* and *f* since, when the model is extended to continuous media, there are an infinite number of oscillation modes. It has to be noticed that, in principle, all the *n*

natural modes should provide the same value for density. The expression (2) shows the simplest relation between the density of the fluid and the free oscillation frequency of the tube. For Coriolis sensors, tubes are forced to oscillate by an external driver and the value of the natural oscillation frequency is obtained in correspondence of a relative maximum of the oscillation amplitude.

2.2 Geometrical and Physical properties of the system

The model reported in expression (2) does allow to determine the density of a flowing fluid only when all quantities are calculated independently and with the necessary accuracy. Some of the terms, can reasonably be evaluated and corrected for the effect of pressure and temperature, as it happens for the volume *V,* but unfortunately K_n can be estimated, starting from thermodynamic and mechanic principles of the continuum physics, only in the case of ideal conditions. In general, this term comes from the solution of partial differential equations describing an elastic material, of a defined shape, satisfying specific thermal and mechanical boundary conditions. Some efforts have been spent to solve the problem mathematically both using analytical [16] and numerical [17] approaches, the latter is usually based on finite element methods of analysis. Despite the complexity of the solutions, it is commonly accepted that the quality of the predictions is strongly influenced by material properties, thermal, mechanical and boundary conditions that cannot be completely controlled during on-line measurements. From a pure metrological point of view, at the moment, the adopted physical models do not allow to use Coriolis mass flow meters as primary instruments for the measurement of the density though their performances are improving and it can be envisioned that they can be used as secondary standard when accurately calibrated in a limited range of conditions.

2.3 Calibration of Coriolis sensor for density measurements

When it is not possible to determine all the parameters appearing in the function $\rho(f_n)$ described in expression (2), a calibration procedure can be used to evaluate them in a small range of the influence quantities. For example, the term *V* is influenced by the temperature *T* and pressure *p* of the fluid and it can be corrected for these effects when temperature and pressure change. However, corrections for the term K_n are much more complicated to be predicted since its dependence on *T* and *p* passes through other thermophysical properties of the fluid and the tube such as fluid speed of sound, viscosity, elastic properties of the tube, mass distribution and so on. Moreover, adopted models indicate that this term also includes the inertia moment of the system when fluid is flowing. In these conditions, the distribution of the fluid mass within the tube changes with the flow rate and Reynolds number and it is to be expected that the produced effects are different for each natural mode (*n*=1, 2, ...). In this frame, a complete calibration in terms of temperature and pressure for many modes is unrealistic but a specific calibration remains possible.

With the only purpose of fixing the ideas, an explicit expression for the coefficients appearing in expression (2) is reported:

$$\begin{aligned} V &= V_0[1+\alpha(T-T_0)+\beta(p-p_0)] \\ K_n &= k_0 + k_1(T-T_0) + k_2(T-T_0)^2 + h_0(q-q_0) \\ k_1 &= c_{1,0} + c_{1,1}(p-p_0) \\ k_2 &= c_{2,0} + c_{2,1}(p-p_0) \end{aligned} \quad (3)$$

where the dependence of the constants of the system on the temperature and pressure are considered. The choice for the expressions of coefficients reflects the recognized effects described in literature [18] but also observed with direct measurements [19]:

1. Temperature and pressure change the properties of the fluid and of the tube;
2. When sensor is calibrated at ambient temperature T_0, and measurements are performed at cryogenic temperature, K_n shows an approximately parabolic behaviour as a function of the temperature;
3. Linear terms for temperature $k_1(T\text{-}T_0)$ and flow rate $h_0(q\text{-}q_0)$, appearing in K, contribute to describe the changes of the moment of inertia of the system due to the different distribution of the mass of the system;
4. The dependence of the calibration coefficients on the pressure is kept linear for the sake of simplicity, however the effects of pressure have been the subjects of dedicated studies developed at NEL [18] and the results obtained provide clear insight on the required corrections.

2.4 Uncertainty budget

Substituting expression (3) into expression (2), nine calibration coefficients come from expression (3) and one from expression (2) which is the value of m. To reduce the number of calibration coefficients to seven, in some cases, values of α and β can be substituted using the thermal expansion coefficient and the isothermal bulk compressibility of the material which the tube is made of. Sometimes this approximation is not possible especially when the sensor is composed by parts of different materials.

To determine the calibration coefficients of the expression (3), measurements of the density of a reference material can be used, in particular, at the arbitrary condition (T_0, p_0, q_0) where expression (3) simplifies to:

$$\begin{aligned} V &= V_0 \\ K &= c_{0,0} \end{aligned} \quad (4)$$

In this condition, calibration coefficients are (V_0, $c_{0,0}$, m) so that, for this system, the single point calibration is not possible. To obtain calibration coefficients, it is necessary to perform a series of measurements at temperature, pressure and flow rate, close to the condition where the sensor will be used and fit the calibration parameters to represent reference values of density.

When calibration at cryogenic temperature is not possible, sensors are calibrated using a reference fluid (i.e. water) at ambient temperature (usually at 20 °C) and at a higher temperature. At NEL, for example, important tests and measurements have been carried out at 20 °C and 36 °C at a pressure of 0.3 MPa [4]. In that case, density measurements have been collected at 8 different flow rates and it has been shown

that an SI-traceable expanded relative uncertainty of 0.05 % (k=2) can be obtained by using modern Coriolis sensor.

Extrapolation, down to cryogenic conditions, of these encouraging results will be accompanied with additional uncertainty since the effects of temperature, pressure and flow rate would not be fully accounted for, as it happens when performing flow measurements [15].

3. Ultrasonic densimeter

The density ρ of a fluid at rest can be formally determined by measuring the speed of propagation of an ultrasonic tone burst w as $\rho = K_s/w^2$, where K_s is the adiabatic compressibility of the fluid. In this frame, while the speed of sound can be measured independently form the composition, K_s can be determined only when the LNG temperature, pressure and composition is known. Considering that LNG is mainly composed by liquid methane, fractions of ethane (up to 10 %) and propane (up to 10 %), it is possible to evaluate that relative change of the adiabatic compressibility by using accurate equations of state implemented by specialized software like Trend [20] or RefProp [21]. With few trials, it is possible to verify that the adiabatic compressibility can easily change by more than ten percent even under small changes of temperature, pressure and composition. So that, a direct use of a simple expression is not suitable.

Alternatively, the relation between speed of sound and density can be exploited involving the acoustic impedance $Z = \rho w$. In this case, density can be obtained when both the speed of sound and the acoustic impedance is measured at the same conditions of temperature and pressure, without the need to know the composition of the mixture. When the measurement is performed with flowing fluids, acoustic impedance is not perturbed until the speed of the fluid approaches that of the sound (supersonic flows). In practical cases, the density measurement is thus intrinsically independent from flow rate.

Ultrasonic densimeter have been already described in literature [22] since '70s when preliminary trials have been attempted. For density measurements, the acoustic impedance is usually measured creating an interface between the incident acoustic wave-packet, travelling in a buffer, and the fluid. In this way, part of the acoustic energy is reflected and part is transmitted into the fluid. Reflection and transmission coefficients can be determined when the acoustic impedance of the fluid and that of the buffer is known. Conversely, if the acoustic impedance of the buffer is known and the reflection/transmission coefficient is measured, the acoustic impedance of the fluid can be determined. The real challenge to implement an accurate sensor is the measurement of the reflection coefficient.

A laboratory sensor can be realized optimizing the buffer specifications in particular, the size, the shape and the material (having established density as a function of the temperature).

Fig. 1: INRiM's prototype of ultrasonic densimeter.

Figure 1 shows a picture of the first prototype developed at INRiM, two further implementations have been necessary to improve the accuracy of the obtained measurements.

The speed of the ultrasonic wave-packets travelling through the buffer can be determined by measuring the time of flight between the emission and reception time after being reflected at the buffer-fluid interface. Besides, the reflection coefficient can be measured by accurate measurement of the amplitude of the emitted and the received signals, for example by using a high speed 12-bit digital oscilloscope. A complete description of the methods of analysis of the sampled signals is out of the scope of this report but it is usually performed by using spectral ratio methods. In this way, the reflection coefficient can be determined with a relative accuracy of 0.1 % or better. The density of the fluid is then calculated using the following working equation:

$$\rho_{\mathrm{f}} = \frac{1-R}{1+R}\frac{w_{\mathrm{b}}}{w_{\mathrm{f}}}\rho_{\mathrm{b}} \tag{5}$$

where indexes "b" and "f" refer to a buffer or a fluid property. Expression (5) shows the acoustic impedance of the buffer $Z_{\mathrm{b}} = \rho_{\mathrm{b}} w_{\mathrm{b}}$ depends only on temperature and pressure, but not on the properties of the fluid, the flow rates or the speed of sound of the fluid.

3.1 Calibration of the ultrasonic densimeter

Though in laboratory conditions the expression (5) is still investigated for optimizing the sensor, for practical purposes, the acoustic impedance of the buffer can be obtained by calibration with reference fluids as a function of temperature and pressure, so that the expression can be rewritten as:

$$\rho_{\mathrm{f}} = \frac{1-R}{1+R}\frac{Z(T,p)}{w_{\mathrm{f}}} \tag{6}$$

where the calibration function $Z(T,p)$ is usually linear with the pressure and quadratic with the temperature for many solid materials. It is worth to notice that the introduction of the calibration function incorporates most of the acoustic effects that are described with difficulties like the flatness of the wave fronts, reflections on the walls of the buffer, parallelism of the source and reflection planes and so on.

At INRiM, a new ultrasonic densimeter has been designed and realized specifically for working at cryogenic conditions. The sensor has been characterized with water in the temperature range of 20 °C and 50 °C. For such a small range, the calibration function is usually linear even if the density shows a significant curvature. The density behaviour is well represented by the variation of the reflection coefficient R and the

speed of sound *w*. An improved calibration, with relative deviations in the order of 0.1 % can be obtained using a quadratic dependence of *Z* from the temperature. This preliminary test confirmed that the acoustic impedance of the buffer rod is weakly coupled to the properties of the fluid.

The measurements collected during the calibration of the sensor are reported in table 1 where deviations from reference values are reported after the sensor calibration. It is worth to notice that the repeatability represents the main contribution to the uncertainty budget as it expected also for other type of dynamic densimeter. By the described calibration procedure it is possible to estimate that the expanded relative uncertainty (*k*=2) of the ultrasonic density sensor is in the order of 0.1 %.

T / °C	R_{exp}	w_{exp} / (m/s)	ρ_{exp} / (kg/m^3)	ρ_{ref} / (kg/m^3)	$(\rho_{exp}-\rho_{ref})/\rho_{ref}\ 10^2$
19.676	0.345151	1481.3	998.26	998.27	0.00
29.654	0.362886	1508.3	996.29	995.75	-0.05
29.652	0.363335	1508.3	995.25	995.75	0.05
39.631	0.381885	1528.3	992.01	992.36	0.04
39.630	0.381600	1528.3	992.67	992.36	-0.03
49.610	0.400760	1542.1	988.22	988.21	0.00

Table 1. Calibration of the ultrasonic densimeter using water as reference fluid in the temperature range of 20 °C and 50 °C. The subscript “exp” indicates measurements obtained in laboratory while “ref” are values obtained using RefProp equation of state [21].

The calibration of the ultrasonic densimeter at cryogenic temperatures can be performed using reference values of density, obtained by primary measurements in pure methane, in the range of temperature between 100 K and 130 K, and for pressure up to 10 MPa, that are the typical conditions where LNG are used. In principle, calibrations on extended temperature ranges are possible but not yet verified experimentally.

Using this approach, the ultrasonic densimeter can be used as a transfer standard, after a calibration in laboratory, to verify other sensors, directly on LNG plants.

3.2 Ultrasonic flow meters for density measurements

It is worth to notice that the measurement scheme adopted for the prototype of the ultrasonic densimeter, is already implemented in modern ultrasonic flow meters and some of the expression (6) quantities are already measured. Thus, it is expected that adapting the expression (6), possibly including also the transmission coefficient, it would be possible to use ultrasonic flow meters to determine the density of the flowing fluid. This innovation would provide a two-fold effects: it could allow to monitor the quality of the fluid and could improve the accuracy of the computation of the flowing mass [23], inferred from the volumetric measurement, to 0.5 % or better. Being based on speed of sound measurements, the ultrasonic densimeter can also be used to verify ultrasonic flow meters directly on plants.

Toward a novel calibration traceability for on line sensor cryogenic fluids

In the frame of density measurements, the efforts spent to reduce the gap between today needs of LNG industries and the services provided by metrological institutes have the benefit to stimulate the investigation of new approaches never adopted before. For example, some of most important realizations are:

1. an innovative primary densimeter, based on the principle of the hydrostatic weighing with magnetic suspension, suitable to characterize both pure cryogenic fluids and complex mixtures [1];
2. a novel transfer standard for density measurements necessary to maintain the traceability between laboratory and on-line density measurements [24];
3. a new facility to calibrate the measured density by on-line flow meters directly with LNG [2, 3];

Described new traceability chain represents the basis on which next innovative solutions can be characterized giving a new chance for the development of new technologies with a significant impact on measurement and control processes of cryogenic fluids.

Recommendations for calibration of on-line LNG density sensors

The work performed in 16ENG09 suggests the following recommendations for the calibration of on-line cryogenic density sensors:

1. When calibrated in a temperature range different from that of utilization, on-line density gauges should be characterized, at least, at 3 different temperatures within an interval of 20 °C. In this way, the *k*-coefficient of expression (3) can be calculated with better accuracy;
2. The calibration should occur in a flow rate range corresponding to the particle speed and fluid Reynolds number of the cryogenic liquid flow. At least three different points should be measured across the flow range of application to be able to calculate coefficients h_0 and q_0;
3. When pressure effects are small, compared to those of the temperature and flow rates, they can be accounted for by estimating the parameters $c_{1,1}$ and $c_{2,1}$ from measurements obtained at different flow rates. In other cases, it is necessary to perform specific measurements at constant temperature and flow rates by changing the pressure only;

Due to inherent heat gain from ambient in cryogenic flows, varying sensor behavior as a function of temperature, and from general flow measurement experience, it is recommended to further corroborate these recommendations by testing LNG density sensors under cryogenic conditions.

SI-traceable density calibration of on-line LNG density sensors can be performed in national metrology or designated institute laboratories at ambient and/or cryogenic conditions. This is recommended when a new type of density sensor or measurement scheme is tested for its accuracy.

On-line density sensors should have a target measurement uncertainty to below (0.46%; k = 2) to be compatible with the current density measurement uncertainty.

On-line density sensors are recommended when cost-effective in terms of CAPEX and OPEX, which could be lower than that of the typically used sampling and composition measuring systems[2], while not compromising on the cost of uncertainty in custody transfer.

References

[1] R. Lentner, M. Richter, R. Kleinrahm, R. Span, *Density measurements of liquefied natural gas (LNG) over the temperature range from (105 to 135) K at pressures up to 8.9 MPa*, Journal of Chemical Thermodynamics **112** (2017), pp. 68-76.

[2] M. D. Schakel, *Liquid nitrogen calibrations of industry-standard LNG flow meters used in LNG custody transfer*", VSL B.V., December 2019, available on line.

[3] M.D. Schakel, et al, *LNG Mid-Scale Loop flow metering – Preliminary Test Results*", Flomeko 2019, 18th Flow Measurement Conference, 26 - 28 June 2019, Lisbon, LNEC, Portugal, 2019.

[4] A. Kenbar, *Measurement of Water Density by Coriolis Flow Meters Under Different Test Conditions,* EMPIR 16EN09 LNG III Report on NEL's water density measurements by Coriolis meter.

[5] G. Cavuoto, S. Lago, P. A. Giuliano Albo, D. Serazio, *Speed of sound measurements in liquid methane (CH_4) at cryogenic temperatures between (130 and 162) K and at pressures up to 10 MPa*, Journal of Chemical Thermodynamics **142** (2020) 106007.

[6] A. Hunt, I. Rusli, M. Schakel, A. Kenbar, *High-speed density measurement for LNG and other cryogenic fluids using electrical capacitance tomography*, submitted to Cryogenics.

[7] A. Kenbar, *Assessment of Calculation Methods for LNG Density Using Available Reference Density Data*, Houston, 2013.

[8] International Group of Liquefied Natural Gas Importers, LNG Custody Transfer Handbook, 3rd Edition ed., Paris: International Group of Liquefied Natural Gas Importers, 2011.

[9] BSI, Refrigerated Light Hydrocarbon Fluids - Measurement of Cargoes on Board LNG Carriers, 1st Edition ed., BSI, 2012.

[2] This is expected to be achievable because Coriolis and ultrasonic flow meters, and the on-line transfer standard densimeter are relatively straightforwardly being built in into cryogenic facilities.

[10] C. Tietz, M. Richter, R. Kleinrahm and R. Span, *Report on the Improvements of the Selected EoS for Calculation of the Saturated Liquid Density of LNG*, EMRP JRP ENG60, 2016.

[11] C. Tietz, M. Richter, R. Kleinrahm and R. Span, *Enhancement of the Revised Klosek and McKinley Method for Density Calculations of Liquefied Natural Gas (LNG) over the Temperature Range from (100 to 135) K at Pressures up to 10 MPa*, Fuel Processing Technology, **165** (2017), pp. 19-26.

[12] O. Kunz and W. Wagner, *The GERG-2008 Wide-Range Equation of State for Natural Gases and Other Mixtures: An Expansion of GERG-2004*, *Journal of Chemical & Engineering Data,* **57**(11) (2012), pp. 3032-3091.

[13] M. Richter, R. Kleinrahm, R. Lentner and R. Span, *Development of a Special Single-Sinker Densimeter for CryogenicLiquid Mixtures and First Results for a Liquefied Natural Gas (LNG)*, Journal of Chemical Thermodynamics*,* **93** (2016), pp. 205-221.

[14] EURAMET, "LNG III: Metrological Support for LNG and LBG as Transport Fuel," 2017. [Online]. Available: https://lngmetrology.info/lng-iii/. [Accessed 30 May 2020].

[15] A. Kenbar, M. Schakel, *Influence of Flow Disturbances on Measurement Uncertainty of Industry-Standard LNG Flow Meters*, Flow Measurement and Instrumentation, (2020), submitted.

[16] S. Guirguis, *Modelling of Coriolis mass flow meter of general plane-shape pipe,* Flow Measurement and Instrumentation **21** (2010), pp. 40-47.

[17] T. Wang, Y. Hussain, *Coriolis mass flow measurement at cryogenic temperatures*, Flow Measurement and Instrumentation, **20** (2009), pp. 110–115

[18] C. Mills, *Calibrating and operating Coriolis flow meters with respect to process effects*, Flow Measurement and Instrumentation **71** (2020), p. 101649.

[19] I. Marfenko, *Long Term Stability of Coriolis Flowmeters in Cryogenic Fluids*, EMPIR 16ENG09 LNG Workshop 2018.

[20] R. Span, T. Eckermann, J. Gernert, S. Herrig, A. Jäger, and M. Thol. *TREND – Thermody-namic Reference and Engineering Data v1.1,2014,* http://www.thermo.rub.de, Lehrstuhl für Thermodynamik, Ruhr-Universität Bochum.

[21] E. W. Lemmon, M. L. Huber, and M. O. McLinden, *NIST Standard Reference Database 23: Reference Fluid Thermodynamic and Trans-port Properties REFPROP, Version 9.0, 2010*.

[22] A. Puttmer, P. Hauptmann, B. Henning, *Ultrasonic density sensor for liquids*, IEEE Transactions on ultrasonics, ferromagnetics, and frequency control **47** (2000), pp. 85-92.

[23] K. Hadidi, A. A. Falnes, *EMPIR 16EN09 LNG III Report on the uncertainty analysis for the LNG flow measurement*.

[24] P. A. Giuliano Albo, S. Lago, G. Cavuoto, *Density Measurements: perspective for traceable on-line sensors*, North Sea Flow Measurement Workshop, 22-24 October 2018, Aberdeen (UK).