



# FINAL PUBLISHABLE REPORT

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## 1 Overview

The overall aim of this project was to enable the large-scale roll-out of liquefied natural gas (LNG) and liquefied biogas (LBG) as transport fuel. The custody transfer measurements of flow, density, and composition need to be underpinned with a clear and traceable metrological infrastructure, and properties which are important for fuel combustion, i.e. the density and the methane number (MN) need to be assessed cost-effectively.

To address this, the project combined expertise from industry, instrument manufacturers, universities, research institutes, and national metrology institutes, to establish the necessary test facilities and validation methods. Key project outcomes are SI-traceable calibration capabilities for LNG flow, composition, and density, and the development of three types of cost-effective MN sensors for LNG engine management. These outcomes of the project were implemented in relevant written standards, and communicated in workshops, conferences, and journal publications to stimulate the use of LNG and LBG as transport fuel.

## 2 Need

As addressed in the "[Clean Transport Fuel Strategy](#)", the utilisation of LNG and LBG as transport fuel constitutes one of the pillars of the European clean fuel strategy. LNG implementation would enable the stringent pollutant emission limits of future EURO VI standards to be met more cost-effectively as compared to conventional fuels. In addition to this, engines running on LNG produce far less noise than diesel-operated engines and are therefore becoming the preferred choice for deliveries in urban areas. LNG is also an attractive fuel to meet the new limits for sulphur content in marine fuels and for nitrogen oxides (NOx)-emissions from ship engines.

The large-scale roll-out of LNG and LBG as transport fuel, however, requires reliable determination of the amount, density, composition and other physical properties of the cryogenic fuel; and although substantial progress had been made in the predecessor LNG metrology projects (EMRP ENG03 and ENG60), important metrological infrastructure and expertise was developed during the project addressing:

*Refuelling/bunkering:*

A calibration facility for LNG flow and composition measurements was constructed at the Rotterdam Port area in the Netherlands. This facility was put into operation with liquid nitrogen (LiN) in 2019 and provides an excellent platform for testing and validation of LNG flow metering technologies suitable for fuelling and bunkering applications. The facility enables systematic research into LNG flow meters under variable cryogenic conditions for which there is high demand from industry.

*Composition, density and LNG particulates:*

The composition of LNG, and consequently the energy content and other physical properties, varies from source to source. Furthermore, the LNG composition in carriers and storage tanks typically changes over time through a process known as "ageing"; which means that the LNG composition gets richer in heavier components. Therefore, standards to accurately define the composition of LNG/LBG mixtures are urgently needed.

LNG produced from Biogas (BG) can contain small particles which affect engine performance over time. Moreover, silicon dioxide particles can be formed through the combustion of siloxanes present in biogas. Therefore, the presence and the source and nature of the particles in LNG from BG also needs to be known to be able to decide the service intervals and the type of particle filters to be used at fuelling stations.

*Engine performance:*

To operate an engine in the most efficient way it should run as close as possible to its knocking point, i.e. the point where the fuel spontaneously ignites. The MN, together with the engine type and the operational conditions, determines this knocking point. Consequently, to run an engine at its most efficient setting, the MN needs to be determined using highly accurate and cost-effective methods.

Depending on the combustion process, part of the methane may not burn in the engine and thus can be released to the atmosphere (also known as methane slip (MS)). Thus, for economic and environmental reasons (i.e. methane is an important greenhouse gas), engine performance needs to be carefully monitored and managed to increase the combustion efficiency and minimise MS.

### 3 Objectives

The goal of this project was to enable the large-scale roll-out of LNG and LBG as transport fuel. The specific objectives were:

1. To reduce the onsite flow measurement uncertainty for small- and mid-scale cryogenic applications to the level comparable to meet the current OIML recommendations (1.5 %). To include a systematic assessment of the impact of flow disturbances and the impact of meter insulation.
2. To undertake a technical feasibility study to develop an LNG flow calibration facility for flow rates typically encountered in small- and mid-scale applications ( $400 \text{ m}^3/\text{h} \sim 1000 \text{ m}^3/\text{h}$ ). The Calibration and Measurement Capability (CMC) of this facility should be low enough to at least meet the current International Legal Metrology Organisation (OIML) recommendations (1.5 %), but ultimately the uncertainty should be comparable to the one for conventional fluids i.e. (0.5 %). Furthermore, to assess whether the (on-site) measurement uncertainty can be reduced using a cryogenic piston prover.
3. To develop and validate a reference liquefaction technique (small scale liquefier) for the validation of LBG and LNG sampling and composition measurement systems.
4. To improve methods and (in-line) sensors for cost-effective measurement of the gas composition, methane number (MN) and methane slip (MS). In particular to: i) develop an SI-traceable density calibration method; ii) validate cost effective (in-line) density sensors; iii) validate sensors for composition and MN to enable real-time engine management, engine performance and the measurement of MS; iv) validate the existing MN algorithm from JRP ENG60 and reaction kinetics through full scale truck experiments; v) assess the source, content and potential impact of particles, particularly in LBG fuels.
5. To facilitate the take-up of the technology and measurement infrastructure developed in the project by the measurement supply chain (accredited laboratories, instrument manufacturers), standards developing organisations (ISO, CEN) and end-users (transport and energy sectors). In particular to: i) input to an ISO standard for cryogenic flow metering, including recommendations on water calibration transferability; ii) input to an ISO standard for the calculation of the MN and iii) implement relevant results from the three LNG projects (EMRP ENG03 and ENG60, and this project) in the International Group of Liquefied Natural Gas Importers (GIIGNL) handbook for LNG custody transfer.

### 4 Results

#### 4.1 *To reduce the onsite flow measurement uncertainty for small- and mid-scale cryogenic applications to meet the current OIML recommendations (1.5 %). To include a systematic assessment of the impact of flow disturbances and the impact of meter insulation.*

In order to investigate onsite LNG flow measurement uncertainty, water calibrations were performed in NEL's UKAS accredited water calibration lab and liquid nitrogen (LIN) calibrations were performed in VSL's LNG research and calibration facility. The same metering lines were used. Currently, Ultrasonic (USM) meters and Coriolis Flow Meters (CFM) are calibrated with water and correction equations are applied to compensate for temperature effects at cryogenic conditions when measuring LNG flow [1]. Due to the delay in commissioning of the mid-scale calibration facility for LNG, it was decided to carry out the cryogenic flow disturbance tests with LIN instead of LNG. The pertinent flow properties of LIN are similar to LNG and therefore this has enabled the study of the meter deviation and impact of flow disturbances under cryogenic measurement conditions and contrasting the meter deviations under ideal and disturbed flow against the water flow calibrations.

At the start of the project, it was planned to investigate the following influencing factors on meter measurement uncertainty under ambient and cryogenic flow conditions:

- Flow disturbances.
- Multi-phase flow.
- Coriolis meter and Electrical Capacitance Tomography (ECT) inclinations.
- Meter insulation.
- Coriolis meter response time, thermal stability, and nonlinearity of Young's modulus versus temperature under cryogenic conditions.

As the test protocol for the calibrations was drafted, it was found that the calibration campaign gained strong interest from the instrument manufacturer stakeholders who donated the LNG meters to the research. Therefore, it was decided to test six LNG flow meters from five different suppliers while not including all testing parameters listed above. Therefore, 2-phase flow and inclination effects were not systematically investigated.

Figure 1 shows an example configuration of the metering line used during the calibrations. Figure 2 shows the LIN calibration facility developed during EMPIR project ENG60, and used to calibrate the metering line of Figure 1.

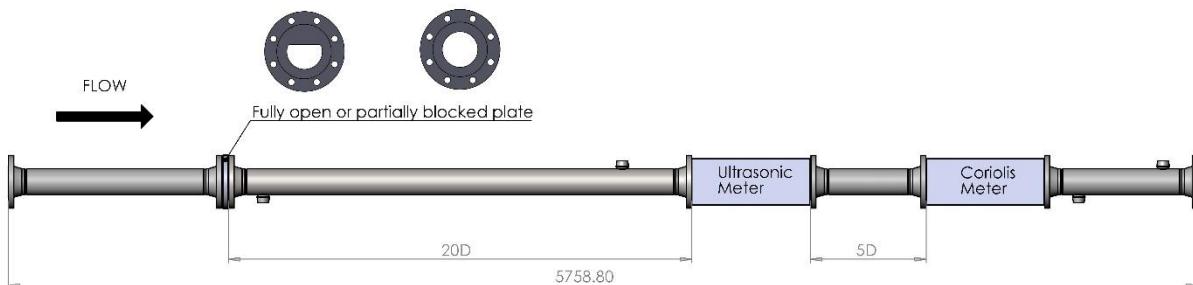


Figure 1: 4" metering line used in water and LIN calibrations of LNG flow meters. Figure taken from [2].



Figure 2: VSL's cryogenic research and calibration facility. Picture taken by Erik Smits, VSL.

The water and LIN calibration results are shown in the publications of the project [1], [2].

From the results of the water and LIN calibration campaign the current status of achievable dynamic LNG flow measurement uncertainty and possible future developments to improve on it was sketched as follows [2]:

"From the results presented in this work and focusing on the test results from the ideal setup, it can be said that the correction models used to transfer the water calibration to cryogenic conditions (using LIN) can potentially result in mass flow rate absolute measurement errors below 0.50 %. Taking into consideration the maximum combined measurement uncertainty of the cryogenic facility of 0.37 % ( $k = 2$ ) for 85 % of the LIN test data, the target measurement uncertainty of 0.50 % ( $k = 2$ ) can be achieved provided that the expanded standard deviation of the mean value, computed from the individual calibration points per set flow rate, is smaller than 0.40 % ( $k = 2$ ). Metrological institutes, meter manufacturers, and LNG custody transfer end-users will continue efforts to (I) make this statement firmer and applicable to LNG, and (II) to reduce the transferability and onsite flow measurement uncertainty to a smaller figure. The present work (water calibration and LIN calibration) is a first important step to assess transferability of meter calibrations with water at ambient conditions to cryogenic conditions. As improvements will be implemented to the system and more industrial experience is gained, the measurement uncertainty will improve to a smaller figure. At this stage, based on the present work, no exact numbers can be stated. This will require more efforts from metrological institutes, meter manufacturers and LNG custody transfer end-users. The end-users will have different uncertainty requirements depending on the application of the flow meter(s), this will dictate whether an ambient calibration (e.g. using water) or a cryogenic calibration (e.g. using LIN or LNG) of the flow meter(s) to be carried out. This is often referred to as "uncertainty tiering"."

Conclusions on the objective are as follows:

1. Absolute meter errors within 0.50 % can be achieved (for Coriolis flow meters) for the cryogenic testing with LIN.
2. The influence of typical flow disturbances on absolute meter error is below 0.50 % when these disturbances are followed by straight piping equivalent to 20 pipe diameters (20D).
3. The LIN calibration results show that the effect of removing the meter insulation on measurement accuracy can be significant (i.e. absolute meter error > 0.50 %).
4. When LNG measuring systems would fall into class 0.5 for type approval (which is not the case at the time of writing), typical for conventional fuels, improved SI-traceable calibration uncertainty is required to be 1/3 or 1/5 of the 0.50 % resulting in required SI-traceable calibration uncertainty of 0.15 % or 0.10 % which at present appears to be a challenging target to achieve.
5. LNG measurement and calibration standards prescribe that boil-off and concomitant 2-phase flow should be avoided. Measuring 2-phase flow accurately is in development phase using a newly developed cryogenic ECT sensor developed by the Coventry University (UCov) and Atout Process Ltd [3].
6. One could conclude that the calibration of LNG flow meters should be performed with a sufficiently small temperature change (i.e.,  $\leq 0.2$  °C/min) for the individual batch run.

A summary of recommendations to reduce LNG flow measurement uncertainty is as follows:

1. To repeat the LIN calibrations with LNG in the same setup as performed in the project to gain more knowledge on meter deviations at LNG process conditions.
2. To perform calibrations in which meter-under-test diagnostic data and cryogenic calibration facility process data are acquired. This will likely help to explain fully and more accurately the larger errors seen in some of the calibrations.
3. To perform calibration campaigns on a varied set of LNG custody transfer meters to arrive at firmer conclusions on transferability of meter calibrations with water at ambient conditions to cryogenic conditions. Currently, this transferability is specific to the meter type, model and manufacturer.
4. Guidelines as prescribed in the standard ISO DIS21903:2020 [4] (developed as part of the project) and the OIML recommendation R117 [5] should be taken into account when performing cryogenic calibrations.
5. The effect of temperature and pressure changes on the measurement uncertainty from line-pack effects in the LNG measurement system should be taken into consideration.

6. For specific flow disturbances of interest (other than those tested in this project), tests should be performed to assess their effect on LNG flow measurement uncertainty and to determine improvement measures to the LNG measurement system at hand.

Using the water and LIN calibration data, JV developed an LNG flow measurement uncertainty model, which helped to firm on the conclusions above.

Initially it was anticipated to include a cryogenic ECT sensor developed by Coventry University (UCov) and Atout Process Ltd [3] into the test campaign. This turned out to be not possible due to the sensor being in development stage. Design challenges relating to electrodes being able to withstand cryogenic temperatures, and selection of suitable cryogenic insulators isolating the electrodes were solved, and the sensor was subsequently tested in LIN [6] under static conditions. Figure 3 shows the LIN-testing setup where the prototype sensor is immersed in a plastic tank filled with LIN.

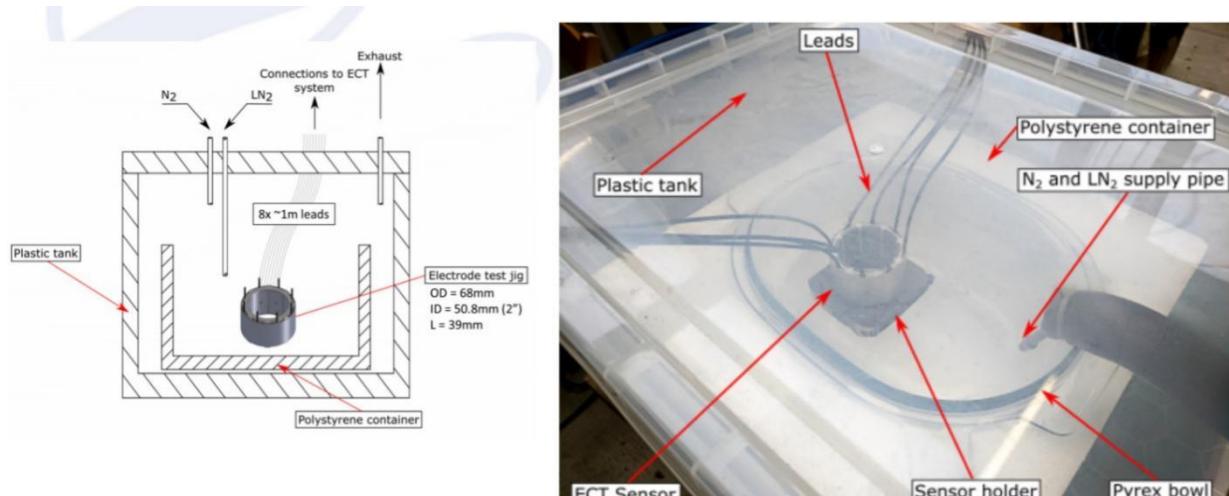


Figure 3: LIN experimental test setup of cryogenic ECT. Figure taken from [6].

From the test results of the newly developed cryogenic ECT sensor the following conclusions were drawn [6]:

1. Design and testing of a single ring ECT sensor have shown proof of principle.
2. Measurement of the relative permittivity of LIN of 1.45 shows a standard measurement error of 0.034.
3. At static cryogenic conditions the root-mean-square variation of the output is better than 0.001 relative permittivity units.
4. The primary errors are associated with the unguarded nature of the test sensor, other errors are identified but not yet quantified.

The proof of principle of the cryogenic ECT is encouraging to the field of the measurement of cryogenic 2-phase flow, as they can provide images of gas in the liquid and the concentration and velocity of the gas bubbles. Achievable measurement uncertainty on these values is currently unknown, but 1 % appears to be feasible [6], [7].

The LIN calibration reference standards are either based on the use of a primary reference system (e.g. a gravimetric system) or based on the use of a secondary reference (e.g. master meters such as Coriolis mass flow meters). An alternative and a novel primary reference principle is based on the use of cryogenic Laser Doppler Velocimetry (LDV). A prototype cryogenic LDV sensor was developed under the predecessor project (ENG60) [8]. During the project it was further improved by Cesame, in collaboration with Engie, Elengy, and Naturgy & Reganosa (experimental support, maintenance handling and accessibility), and CMI (CFD for LDV nozzle optimization and investigation). Improvements, amongst others, were made to its seeding unit and its safe use in potentially explosive atmospheres [9]. Figure 4 shows pictures of the improved cryogenic LDV.

The cryogenic LDV can be used to scan the radial velocity profile or to perform a local velocity measurement along the radial profile and infer the volume flow rate from it. The first mode corresponds to a primary technique while the second mode is faster and currently corresponds to a secondary standard [9]. The relative standard uncertainty is flow dependent. The shape of the dependence can be recognised from the in-use uncertainty of the calibration function  $A(Re)$ , plotted in Figure 5.

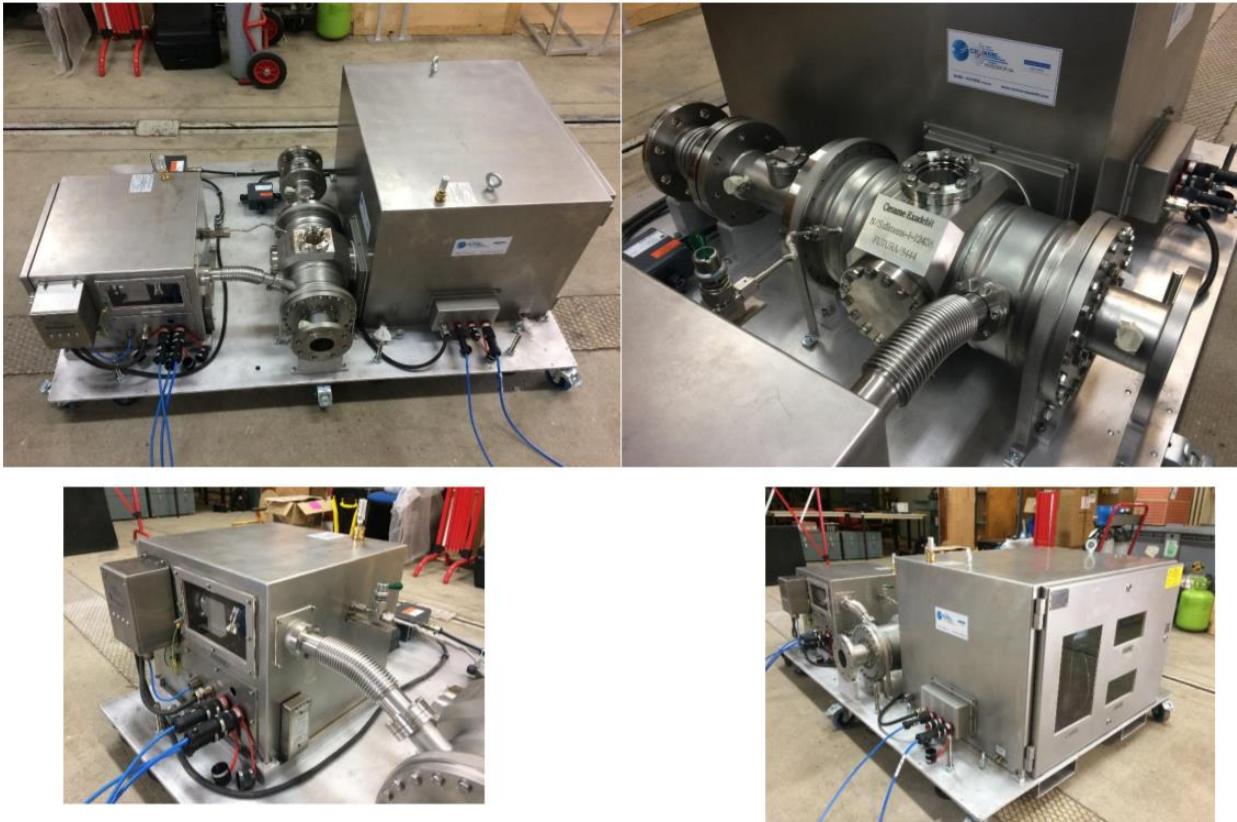


Figure 4: pictures of improved cryogenic LDV. Pictures taken from [9].

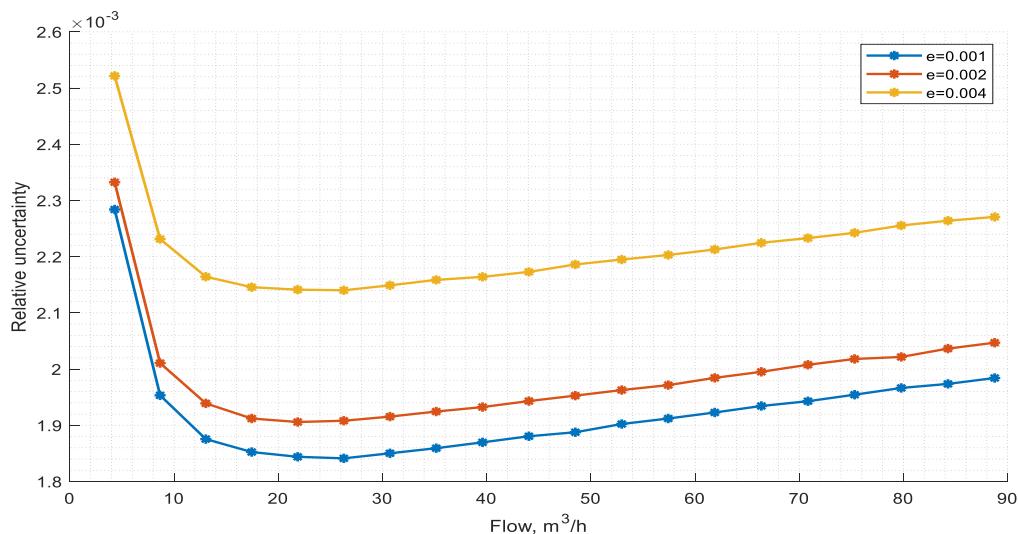


Figure 5: Standard uncertainty for measurements ranging from 1 m/s to 20 m/s with inputs outlined in input data section. The graph shows three different values for the model error  $e$ , to indicate the influence of changes to this input. The expanded uncertainty is not shown, but the coverage factor is very close to 2.

The improved cryogenic LDV was tested further in two test campaigns. The first test campaign took place at the Reganosa LNG terminal and consisted of a weighing bridge, Coriolis flow meter and the LDV standard (Figure 6). Compared results of totalized masses of a LNG batch showed that the weighing bridge and cryogenic LDV results were in agreement to within 0.6 % ( $k = 2$ ) after correction for the boil-off gas, which could be independently measured in the experiment [9]. The second test campaign took place at the LNG terminal in Montoir. Results of the flow meter and the cryogenic LDV deviated by about 2.6 %. In this case, no direct comparison with the weighing bridge could be made.

It is foreseen that the cryogenic LDV can be used as a reference for cryogenic flow meter calibration or as an alternative measurement system to Coriolis and ultrasonic flow meters [9].

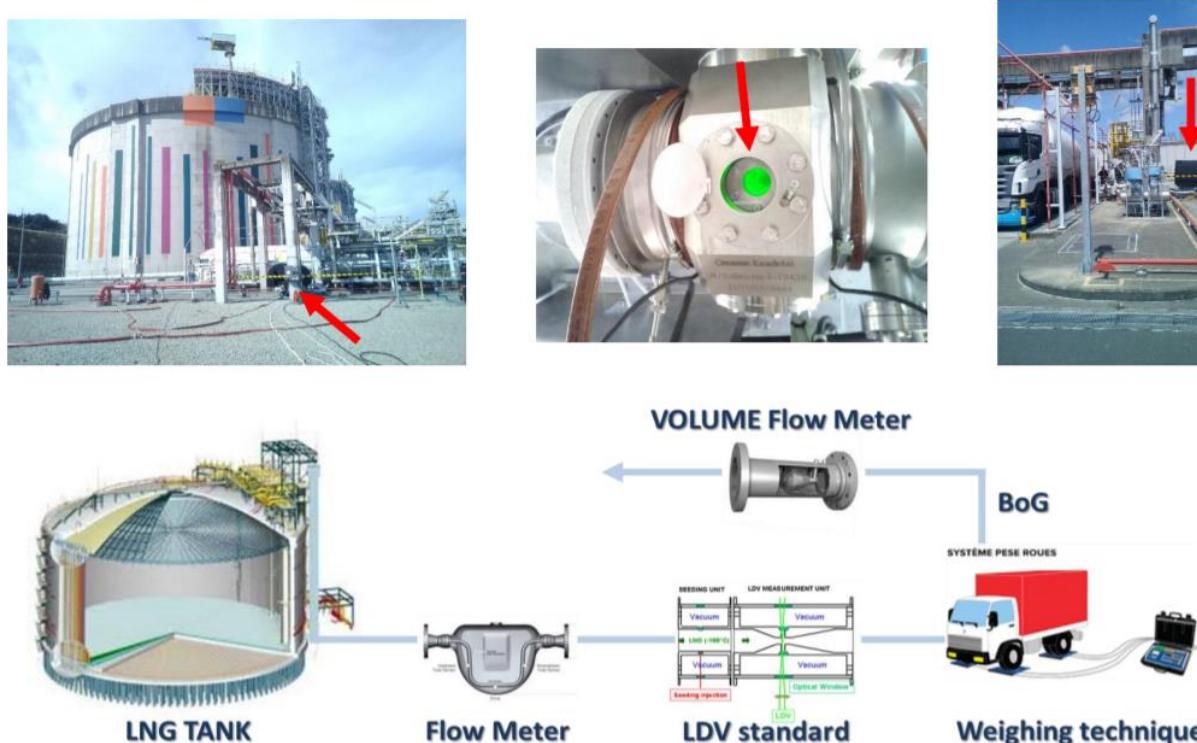


Figure 6: schematic of cryogenic LDV validation at an LNG terminal. Red arrow indicates location of the cryogenic LDV. Figure taken from [9].

In summary, the key outputs relating to the objective are:

- Cryogenic research and calibration facility for LIN calibrations of LNG meters and the comparison of water and LIN calibrations in different typical LNG installation flow geometries.
- Proof of principle of cryogenic ECT shows potential use for LNG flow measurement, including 2-phase flow.
- Validation of cryogenic LDV as alternative primary measurement technique for LNG flow measurement.

Given the uncertainties stated above, the research objective was achieved.

**4.2 To undertake a technical feasibility study to develop an LNG flow calibration facility for flow rates typically encountered in small- and mid-scale applications ( $400 \text{ m}^3/\text{h} \sim 1000 \text{ m}^3/\text{h}$ ). The CMC of this facility should be low enough to at least meet the current OIML recommendations (1.5 %), but ultimately the uncertainty should be comparable to the one for conventional fluids i.e. (0.5 %).**

Figure 7 shows the global distribution of the LNG custody transfer market. The small- and mid-scale applications ( $400 \text{ m}^3/\text{h} \sim 1000 \text{ m}^3/\text{h}$ ) correspond to its use as transport fuel for buses, trucks and ships. Typical flow meters used in this range are USM's and CFM's, where USM's are also typically used as process flow meters at large scale (i.e.  $> 1000 \text{ m}^3/\text{h}$ ) custody transfer applications.

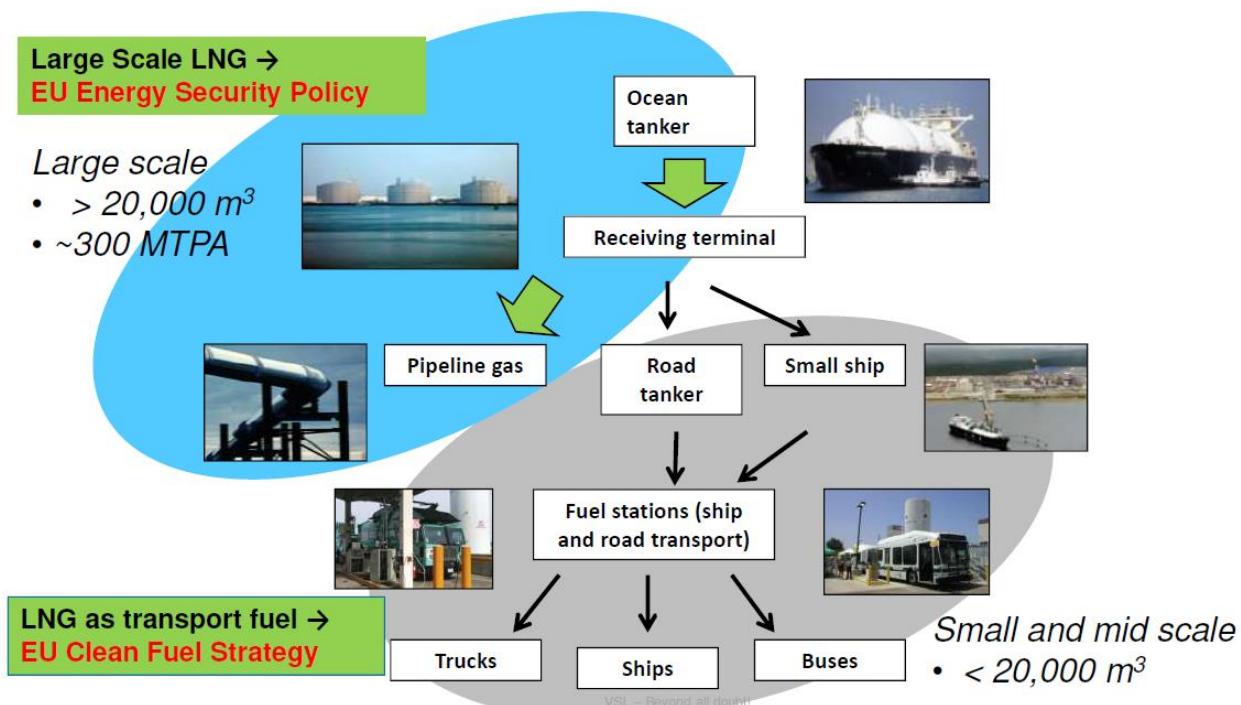


Figure 7: LNG distribution scheme. Numbers estimated in 2018. Figure taken from [10].

Figure 8 shows an USM for measurement of large LNG volumetric flow rate of LNG at an LNG facility. This project determined a practical SI-traceable USM flow measurement uncertainty under cryogenic circumstances for flow rates up to  $100 \text{ m}^3/\text{h}$  by performing calibrations with LIN on two 4"-meters [1], [2] as described in 4.1. Further work would be required to cover the full small to large scale range (USM's with diameters up to as large as 50"). Note that larger meter diameters up to 50" sizes are currently used for allocation or non-fiscal measurements.



Figure 8: 44" USM used in LNG facility. Picture used with permission from Panametrics, a Baker Hughes business.

Two measurement uncertainties are stated in the objective: 0.5 % and 1.5 %. For metering system type approvals, typically prescribed SI-traceable calibration uncertainty is 1/3 or 1/5 of the maximum permissible error, depending on the class. For a class 1.5 measuring system currently applicable to LNG [5], i.e., having a maximum permissible error of 1.5 %, the SI-traceable calibration measurement uncertainty should not exceed 0.5 %, which is 1/3 of the maximum permissible error. Within the project, (provisional) primary standard measurement uncertainties of 0.30 % [1] and 0.60 % [8] were stated. In order to enable class 0.5 calibrations, smaller measurement uncertainties of the primary standards are required. The project has progressed beyond the current state of the art by providing validated primary standards for small-scale LNG custody transfer flow measurements and class 1.5 LNG metering system type approvals.

The initial design of the LNG research and calibration facility, which was made under predecessor project ENG60 and validated for LIN flow rates up to 100 m<sup>3</sup>/h in this project, included a third line to expand the targeted flow rate to 400 m<sup>3</sup>/h. Figure 9 shows the principle of establishing SI-traceable mass flow rate measurements for the VSL's LNG research and calibration facility. Master meters (FT 117 – FT 120) in the primary standard loop (PSL) are calibrated against the weighing scale. The PSL master meters are then used to calibrate the working standards (FT 101 – FT 104) in the mid-scale loop (MSL). The MSL working standards are then used for calibrating cryogenic flow meters which are installed in the Meter-under-Test (MuT) section. By parallelizing flow lines in the master meter/working standard skids, metrological traceability can be upscaled to higher flow rates, at the cost of an increase in measurement uncertainty. In Figure 9 there are two lines in the PSL, and four lines in the MSL Master Meter (MM) cold box. By inclusion of additional lines, higher flow rates can be reached for calibrations.

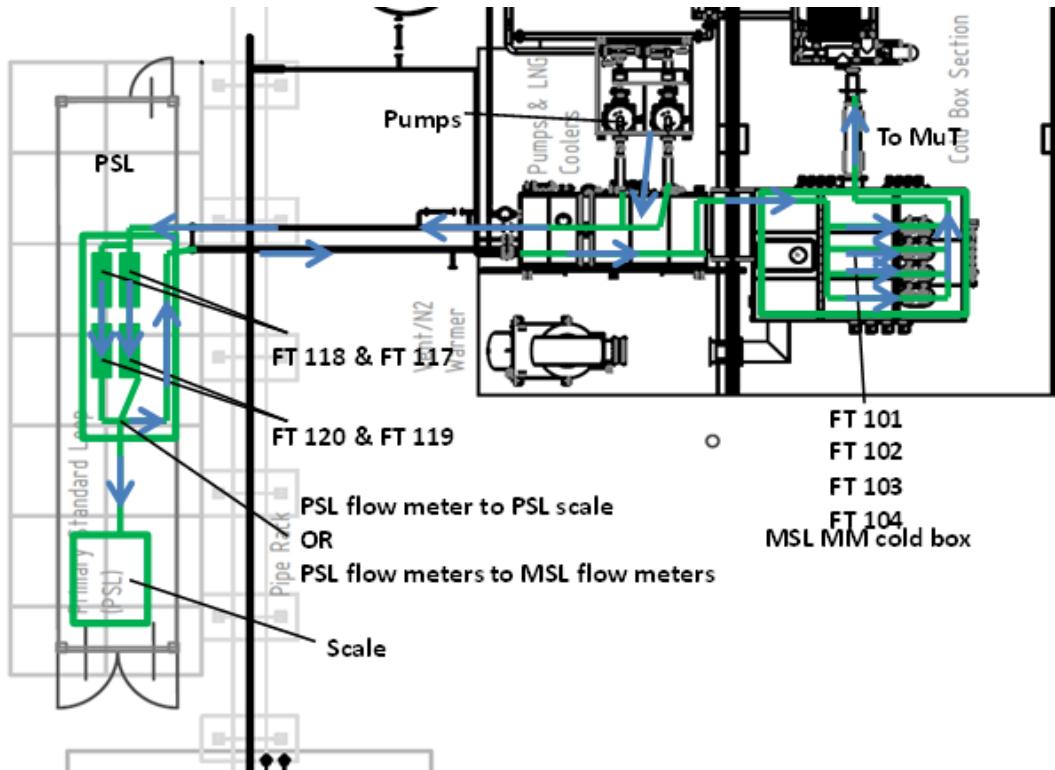


Figure 9: schematic of the LNG gravimetric primary standard and the upscaling method. Picture taken from [11].

The uncertainty in flow calibrations when expanding to higher flow rates was further investigated as part of a cryogenic piston prover feasibility study. The cryogenic piston prover primary standard provides metrological traceability to volumetric flow, while that of the current facility described in Figure 9 is in terms of mass flow. Thus, the facility provides the potential means to compare various cryogenic flow metering principles. It was found that when combining dominant uncertainty sources of the prover and its integration into the facility, the expected lower limit on the expanded uncertainty is about 0.2 % in volumetric flow rate. The study considered a flow rate of 600 m<sup>3</sup>/h fitting well into the range for small- and mid-scale LNG custody transfer applications (400 m<sup>3</sup>/h ~ 1000 m<sup>3</sup>/h).

The cryogenic LDV standard (see section 4.1) was tested and validated with LIN and LNG for flow rates up to about 100 m<sup>3</sup>/h. It is expected that it can also provide calibrations at flow rates covering the small- and mid-scale applications (400 m<sup>3</sup>/h ~ 1000 m<sup>3</sup>/h). As mentioned in 4.1, the cryogenic LDV can be operated as a primary standard or as a secondary standard [9], and its extended measurement uncertainty was determined and estimated to be lower than 0.6 % [8]. When the cryogenic LDV would be used at higher flow rates, one could think of a setup as sketched in the bottom of Figure 6 where the cryogenic LDV is directly connected to the LNG terminal, and the flow rates are higher to correspond to the small- and mid-scale applications.

In summary, the key outputs relating to the objective are the cryogenic piston prover feasibility study and the initial design of the LNG research and calibration facility. At the start of the project, another feasibility study for expanding the flow rates of the calibration facility was planned, which was not completed because more resources were devoted to the objective 4.1 activities. From the cryogenic piston prover feasibility study, the validation of the cryogenic LDV, and the establishment of the LNG research and calibration facility it can be concluded that, given the stated uncertainties, extending these techniques to the small- and mid-scale applications, is possible (400 m<sup>3</sup>/h ~ 1000 m<sup>3</sup>/h), with which the objective was achieved.

#### **4.3 To develop and validate a reference liquefaction technique (small scale liquefier) for the validation of LBG and LNG sampling and composition measurement systems.**

A review of small-scale liquefaction techniques was performed; it included an existing commercially available liquefier from the collaborator EffecTech UK, which is accredited to ISO/IEC 17025 by UKAS, the UK accreditation service. From the review it was found that the supercritical liquefaction technique developed for the densimeter built at RUB [12] in preceding LNG metrology projects (ENG03 and ENG60) could be adopted to a reference liquefier, and a corresponding modified design was developed [13]. Figure 10 shows the design, in which the measuring cell, vapor liquid equilibrium (VLE) cell, and composition measurement (pilot) probe are indicated. However, it was anticipated that building this liquefier with its supercritical liquefaction technique would be too costly, and therefore it was not built as part of the present project.

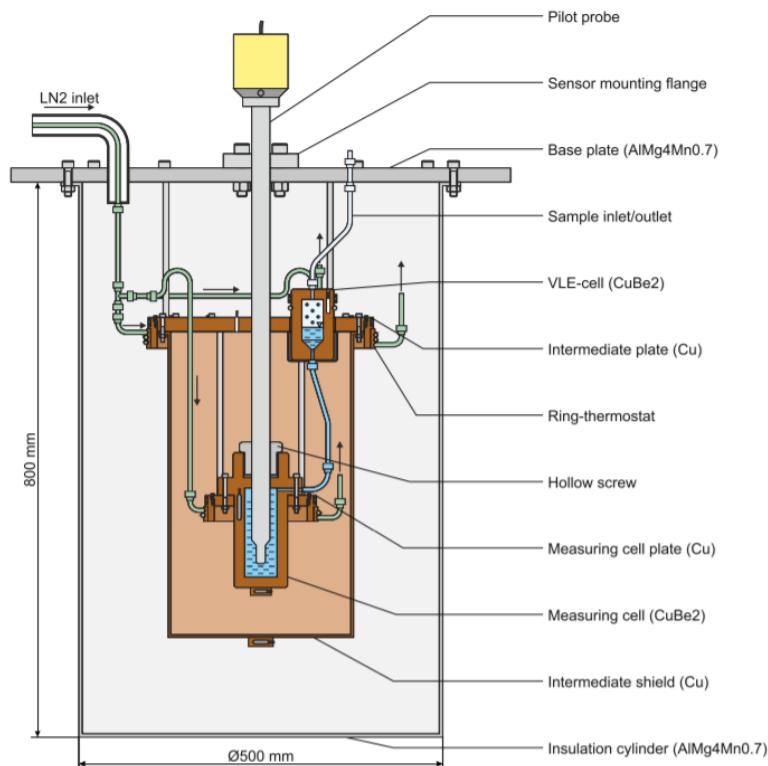


Figure 10: Schematic illustration of the conceptual design for a small-scale reference liquefier with installed optical Raman probe; figure taken from [14].

Hence, a collaboration with EffecTech UK was established to perform metrological tests with the commercially available liquefier, which utilizes condensation as liquefaction technique. Figure 11 shows a picture of the EffecTech Cryostat. Performance tests were carried out with this cryostat using VSL's certified gas mixtures with components normally found in LNG. The test results were compared with the thermodynamic characterization of the liquefier using state-of-the-art equations of state (EOS) for LNG, as implemented in the TREND 4.0 software made available by RUB within the scope of the project. This work showed that the EffecTech liquefier can be successfully employed for metrological purposes, such as the calibration and performance evaluation of optical sensors for LNG-composition measurement. The uncertainty of the liquid composition is comparable to state-of-the-art calibration capabilities for natural gas. An example is given in Table 1. A VSL certified reference gas mixture went through a condensation/liquefaction process, was pressurized, sampled, vaporized and then sent to a gas chromatograph for analysis. Based on the GC analysis, a correction for the methane and nitrogen content was applied to the measured liquefied sample. Table 1 shows the final composition measurement and its uncertainty. This composition is metrologically traceable to internationally accepted standards for natural gas composition.

<b>component</b>	<b>Amount fraction (%)</b>	<b>Uncertainty (k = 2)</b>
nitrogen	0.7884	0.0073
methane	86.9362	0.0350
ethane	7.0474	0.0135
propane	3.0329	0.0092
iso-butane	0.9984	0.0035
n-butane	0.9985	0.0035
iso-pentane	0.0975	0.0007
n-pentane	0.1008	0.0007

Table 1. Final (corrected) liquid composition measurement of a liquefied reference gas mixture and the corresponding uncertainties.

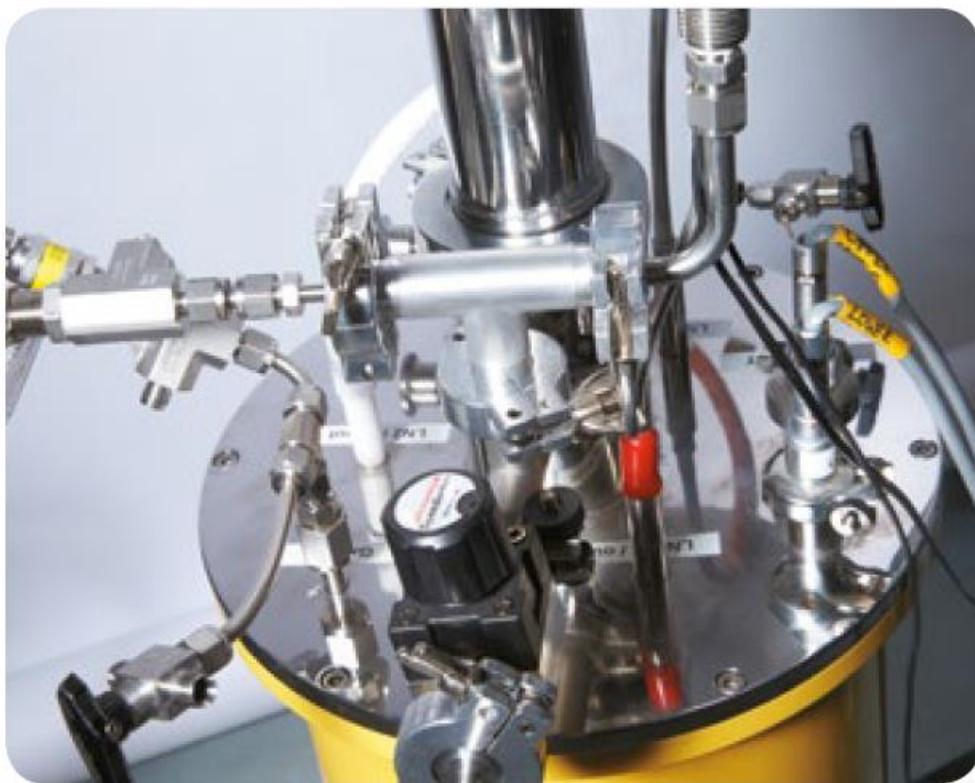


Figure 11: Picture of the condensing cryostat for LNG which is accredited to ISO17025 by UKAS. Picture taken with permission from EffecTech brochure [15].

As part of the project, also density measurements of four liquefied gas mixtures were carried out; here, two methane-rich binary hydrocarbon mixtures and two biogas-like mixtures were studied. Figure 12 shows a

photograph of the cryogenic densimeter developed at RUB that was used for this project. At the center is a rugged frame with the core apparatus (housed inside a large insulation cylinder) that is mounted underneath the base plate; the analytical balance required for gravimetric densimetry is installed at ambient conditions above the base plate under a draft hood. Measurement electronics (pressure, temperature etc.) are placed to the left, and the valve plates for process control (coolant, sample inlet/outlet) are on the right. Figure 13 shows the principle of supercritical liquefaction in a pressure vs. temperature phase diagram (green path in Figure 13). The objective is to measure the density in the homogenous cryogenic liquid mixture inside the measuring cell with traceability in composition to the gravimetrically prepared reference gas mixtures. The sample is liquefied by filling the system composed of measuring cell (M), vapor-liquid equilibrium cell (V) and pressure measurement circuit (P) at ambient temperature ( $T \approx 293$  K); the target pressure is set to a pressure that is significantly higher than the phase boundary indicated by the black envelope. Subsequently the system (M and V) is cooled down while the pressure is measured at ambient temperature (as the precision pressure transducers are installed at the valve plate and controlled at constant temperature). The measuring cell is cooled down to the temperature of interest. Once this temperature is achieved first density measurements in the supercritical region can be performed. To reach the next state point along an isotherm, sample is vented. At some point, a vapor-liquid equilibrium occurs inside the VLE-cell (V). From this point on, the pressure is controlled via the temperature of the VLE-cell; no sample is vented any more. During the present project, an adjusted supercritical liquefaction technique was developed where the VLE-cell remains in the supercritical or homogenous liquid region during the entire investigation [16, 17] (blue path in Figure 13). This is advantageous over the previous technique as it drastically reduces the measurement efforts and, therefore, clearly improves the overall measurement procedure.



Figure 12: Photograph of the cryogenic densimeter set up at the thermodynamics institute of Ruhr University Bochum. Picture taken from [16].

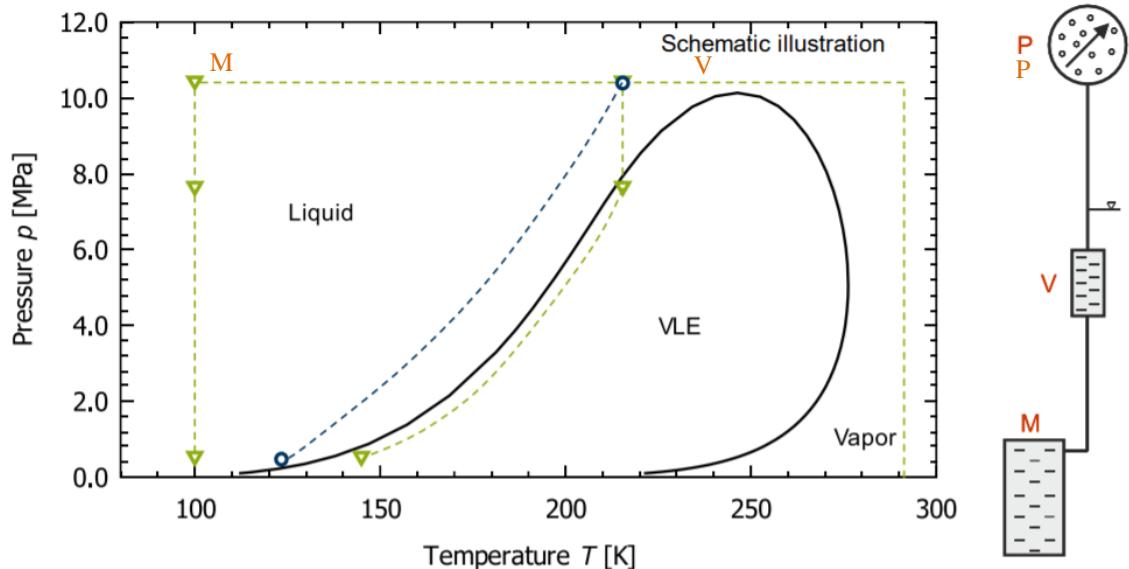


Figure 13: Schematic showing the principle of supercritical liquefaction. Diagram modified from [16].

The experimental liquid densities measured with the densimeter at RUB were compared to state-of-the-art equations of state (EOS). Figure 14 shows an example for a liquefied binary gas mixture consisting of methane and iso-pentane. The GERG-2008 EOS [18] shows relative deviations of up to 2.2 %, which is outside the stated uncertainty of the EOS. In contrast, the EOS LNG [19], developed in a preceding LNG metrology project (ENG60) reveals deviations within the range of (-0.101 to -0.164) %, which is within the estimated uncertainty of this new model. The new experimental data, in particular those for the binary mixtures, will serve to further improve multi-parameter mixture EOS.

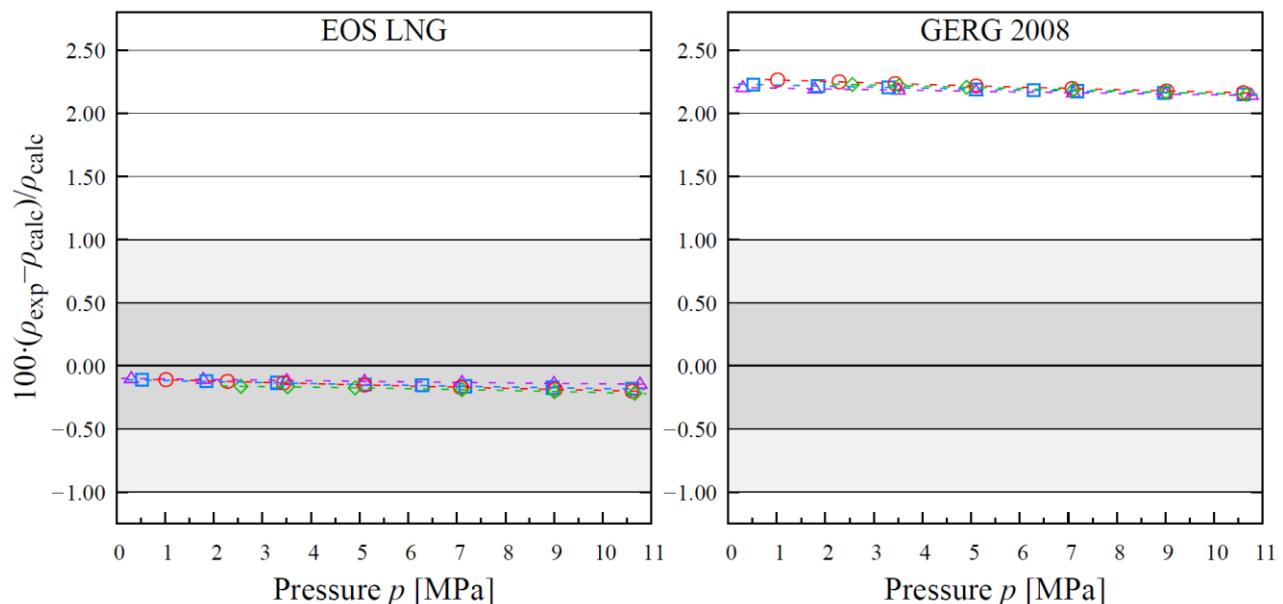


Figure 14: Relative deviations of experimental densities  $\rho_{\text{exp}}$  from calculated densities  $\rho_{\text{calc}}$  for the binary (methane + iso-pentane) mixture. Densities calculated with the EOS-LNG [19] (left); densities calculated with the GERG-2008 [18] (right).  $\triangle$ , 100 K;  $\square$ , 120 K;  $\circ$ , 140 K;  $\diamond$ , 160 K. Graphs and data taken from [16].

In summary, the key outputs relating to the objective are:

- A review of small-scale liquefaction techniques was performed.
- Collaboration with EffecTech UK and the consortium showed that the EffecTech liquefier (accredited to ISO 17025) can be successfully employed for metrological purposes.
- Design of reference LNG liquefier based on a supercritical liquefaction technique realized.
- Density measurements of four liquefied reference gas mixtures with primary cryogenic densimeter show improved match with EOS LNG.
- Improved EOS for LNG thermodynamic property modelling embedded in (amongst other EOS) Trend 4.0 software which is available to the public.

Consequently, validation of a reference liquefaction technique and development of a supercritical liquefaction technique for the generation of reference data relevant to LNG was achieved meeting and exceeding the research objective.

#### **4.4 To improve methods and (in-line) sensors for LNG density and cost-effective measurement of the gas composition, MN and MS.**

The prototype LNG density and speed-of-sound measurement device that was produced by INRIM in the preceding ENG60 project was validated for the measurement of cryogenic speed-of-sound and the results were publicized [20]. LNG density measurements can be performed directly (no need for composition measurement and computation of density from an equation of state) with the device. The measurement principle is illustrated in Figure 15 and is based on measurements of the fluid speed of sound and reflected waves. As the reflections are sensitive to the (LNG) fluid density, a direct measurement of it can be made. During the project, the direct measurement of both LNG speed-of-sound and LNG density was started under static fluid conditions at INRIM. SI-traceable LNG density measurements were performed with the RUB densitometer (see section 4.3) which can be used to validate the INRIM sensor. In parallel, SI-traceable water density measurements by CFM's were performed at NEL under different test conditions as part of the measurement campaign described in 4.1, and it was found that absolute water density errors were within 0.2 % (established with 0.03 % ( $k = 2$ ) combined measurement uncertainty).

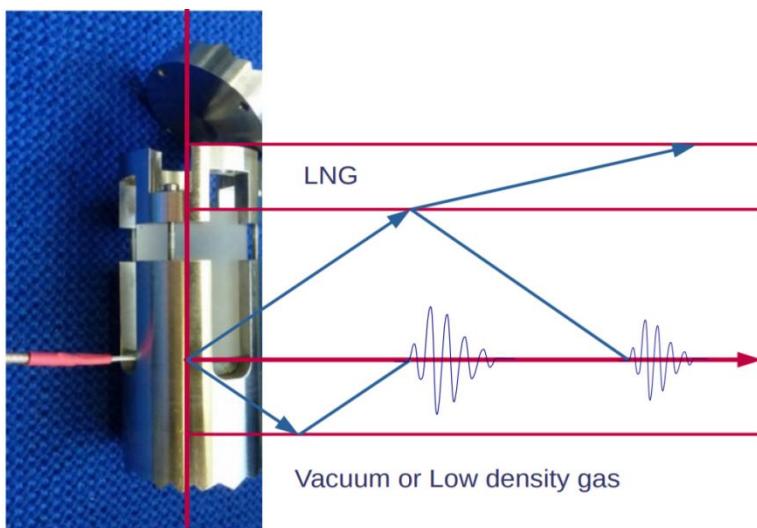


Figure 15: Cryogenic speed-of-sound and densitometer (first prototype) developed by INRIM enabling direct on-line LNG density measurement. Picture taken from [21].

The methane number (MN) of an LNG fuel is a measure for the combustion performance, just like the Octane Number for petrol engines. Unfortunately, there is no direct analytical relationship between the fuel composition and the MN. The MN parameter must be measured experimentally, using a well calibrated engine. By assessing the knocking behavior of different gas mixtures and comparing these with actual fuels, the experimental MN is obtained. To enable the calculation of the MN from the composition, and not only rely on experimental engine tests, much research has been conducted to calculate or approach the MN using analytical models. Examples of such models are Anstallt für Verbrennungsmotoren (AVL), Motorenwerke Mannheim (MWM), National Physical Laboratory (NPL), and Wärtsilä (WMN) [22].

In the project, three gas sensor principles were developed for the measurement of the composition and MN of gasified LNG:

1. Electro Chemical Capacitive sensor array (ECC)
2. Tunable Filter Infrared (TFIR)
3. Fourier Transform Infrared (FTIR)

The sensors are illustrated in Figure 16 (ECC), Figure 17 (TFIR), and Figure 18 (FTIR), respectively.

The ECC sensor is composed of 8 – 10 chips to measure changes in the capacitance values. Each of the chips is coated with a gas absorbing coating. The absorption of gas changes the capacitance value [23].

The TFIR sensor is composed of a tunable Fabry-Perot interferometer with light source, a temperature stabilized sample cell with an absorption length of 40 cm and optical fibres to connect the optical parts [23].

The FTIR method combines the FTIR spectrometer displayed in Figure 18 with a multivariate regression technique. The development of the method involved training to relate the IR-spectra to the MN's by the regression technique [22].



Figure 16: Electro Chemical Capacitive sensor array (ECC) for MN measurement developed by TNO. Picture taken from [23].

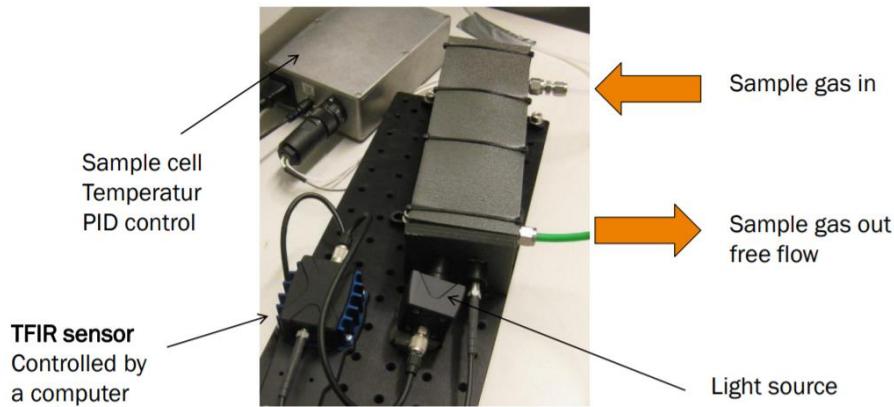


Figure 17+: Tunable filter infrared sensor (TFIR) for MN measurement employed by VTT. Picture taken from [23].

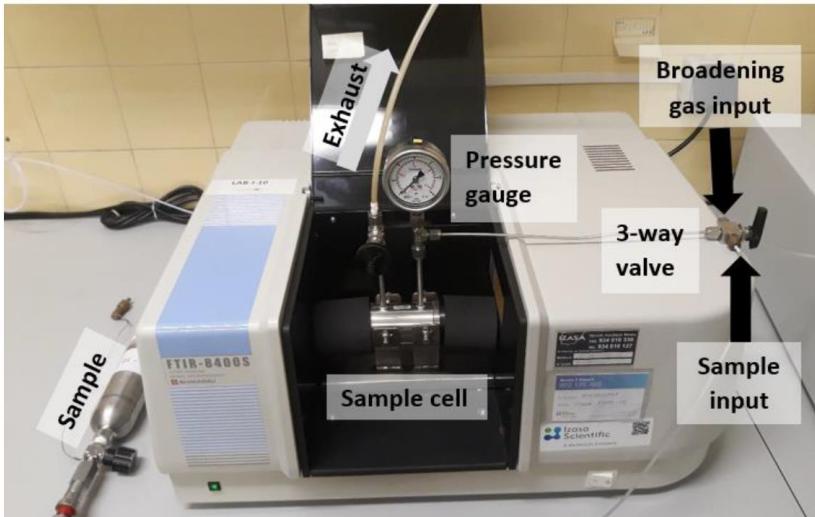


Figure 18: FTIR spectrometer setup. The FTIR method was developed by Naturgy, Reganosa, and Mestrelab with support from the University of A Coruña. Picture taken from [22].

At the beginning of the project, the requirements for the methane number detection were defined, including the type of detectable components, operating temperature and pressure, response time, and MN accuracy. The three sensor methods were bench-marked in laboratory tests against the requirements using sixteen reference gas mixtures provided by TUBS comprised of hydrocarbons C1 – C5 (i.e., methane, ethane, propane, butane, and pentane) and their isotopomers which formed a common, traceable reference set. Figure 19 shows validation results for the ECC and TFIR sensor methods. When cross-correlating the MN prediction from the sensor with the direct determination of the MN in engine tests (ECC) or the computed value obtained directly from the known composition from the Wärtsilä Methane Number algorithm (TFIR), ideally, all measurements will plot along the  $y = x$  line.

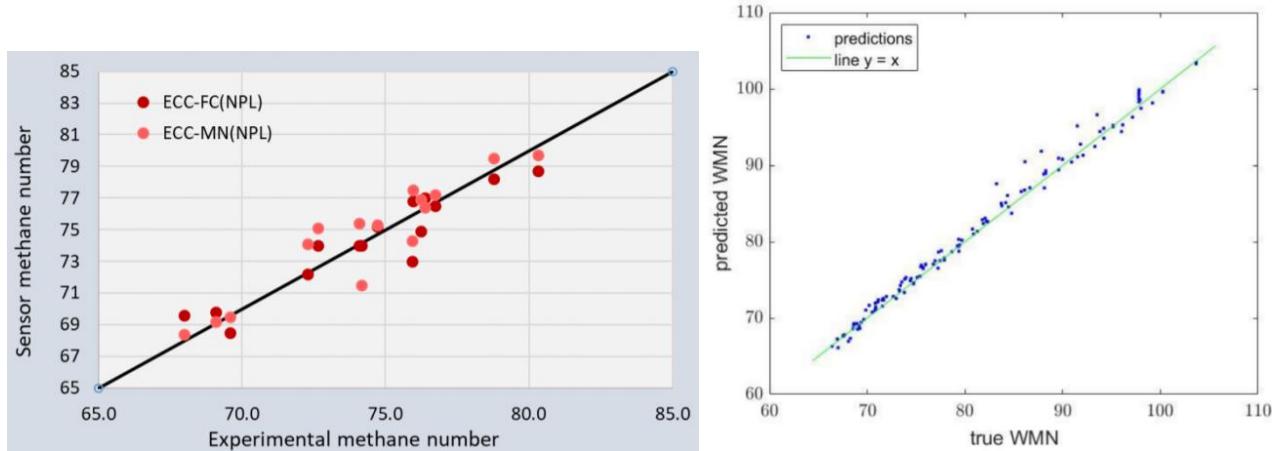


Figure 19: ECC methane number versus reference methane number determined in engine tests by TUBS and PTB (left) and TFIR methane number versus reference methane number (right) computed directly from the composition. NPL and WMN refer to NPL and Wärtsilä MN computation methods respectively. FC and MN refer to (methane number calculation) from „full composition“ and from ECC sensor measurements directly, respectively. Results taken from [23].

In Figure 20 measurements of the MN from the ECC, TFIR, and FTIR methods are compared against the reference value as determined in engine tests. All measurements (sensor and engine tests) used the same set of reference gas mixtures with known composition. From the figure the following statements can be made:

1. All detectors/sensors measure the MN within 1.5 units standard deviation.
2. The MN standard deviation (within 1.5 units) is comparable (given the selected MN algorithm and measurement method).
3. The TFIR method underestimates the MN when compared to the experimental service MN (determined in engine tests, see below).
4. The FTIR method overestimates the MN's.
5. The TFIR method is more accurate for the lower MN's.
6. The FTIR method is more accurate for the higher MN's.
7. The ECC method shows more constant deviations over the whole MN range.

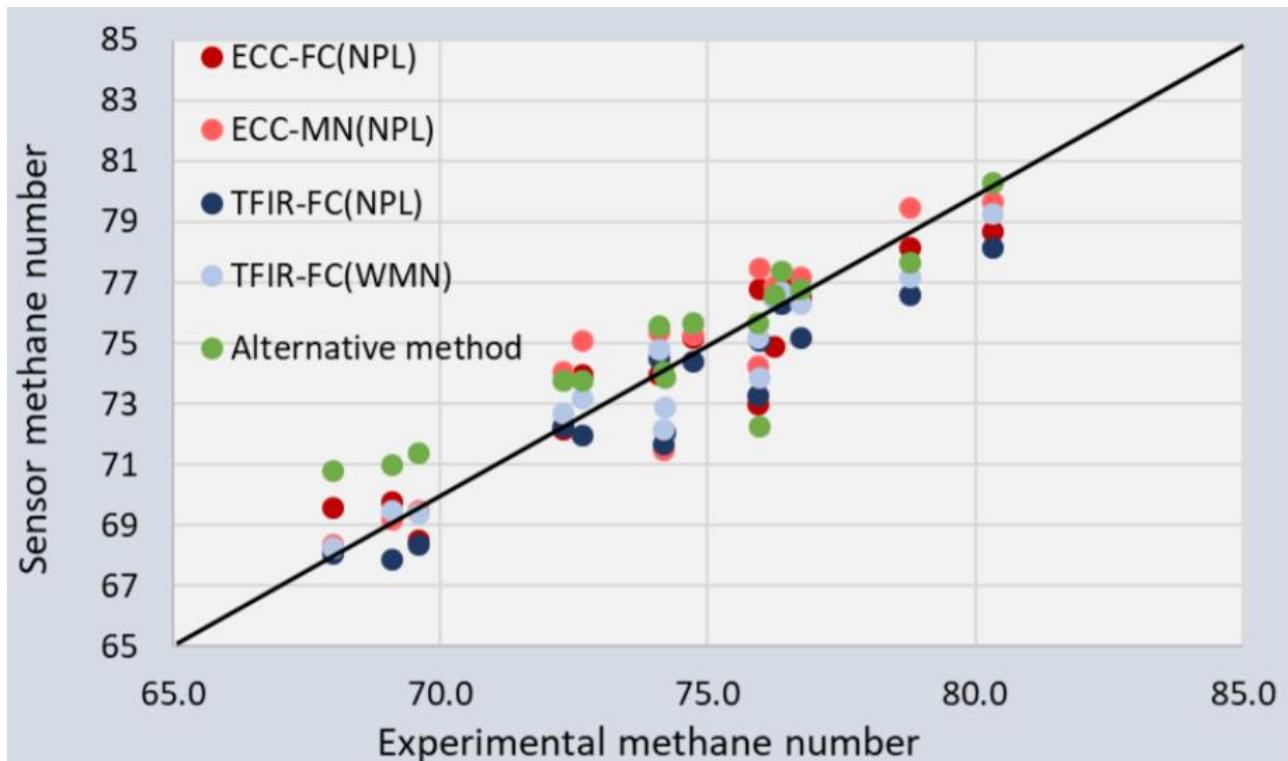


Figure 20: ECC (red, pink), TFIR (blue, lighter blue), and TFIR (green and referred to as alternative method) MN measurements versus reference service methane number as determined in engine tests. FC and MN refer to (methane number calculation) from „full composition“ and from ECC sensor measurements directly, respectively. Results taken from [23].

At the end of the project the sensors were compared in terms of the application parameters which is displayed in Table 2.

## APPLICATION PARAMETERS

Parameter	ECC	TFIR	Alternative method
Standard deviation	1.2	1.0	1.5
Measured parameters	Composition, MN	Composition, MN	MN
Additional parameters	WI, CV, density	WI, CV, density	-
Response time	10-30 minutes	< 10 seconds	1-5 minutes
Position	In-line in fuel line	Off-line in engine bypass	Off-line in lab or bypass
Applicable without T, P control	Yes	No	No
Size	~20 cm <sup>3</sup>	~1000 cm <sup>3</sup>	Lab-equipment
Costs	1-2 k€	2-5 k€	40 k€

Table 2: Comparison of ECC, TFIR, and FTIR (indicated by „alternative method“) sensor methods for the determination of the MN. Table taken from [23].

From Table 2, the following conclusions and recommendations can be made [23]:

- The ECC sensor method is smaller and lower cost than the IR based solutions and requires no temperature and pressure control. However, the response time is longer, which makes the current prototype not very suitable for direct motor management. Additional manufacturing process control and miniaturization will enable faster responses.
- The TFIR sensor method is a fast technique at reasonable costs, but it cannot be directly implanted in a fuel line. When temperature and pressure dependency parameters are accounted for, no temperature and pressure control is required anymore, which may result in in-line applications.

- The FTIR sensor and correlation method is a reliable and robust laboratory technique for gas analysis. Compared to gas chromatography, the technique is lower in cost. It is predicted that this tool will only be deployed in major LNG facilities, not in fuel stations, or car engines.
- All analysers can be deployed as fuel quality monitors for LNG engine management.

The ECC sensor was used for an assessment study towards the suitability for in-line monitoring of the LNG fuel composition and Methane Number. The experiments were conducted at TNO Powertrains in Helmond, the Netherlands, using a 6 cylinder dual-fuel truck engine with a rated power of 250 kW. Figure 21 shows the ECC installation in the engine test setup.

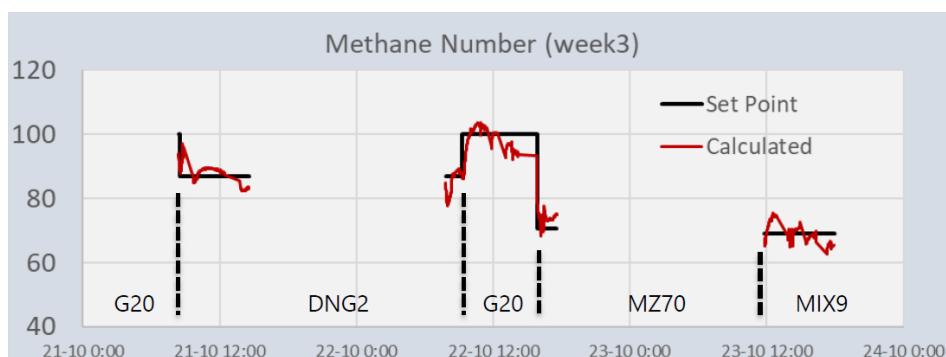
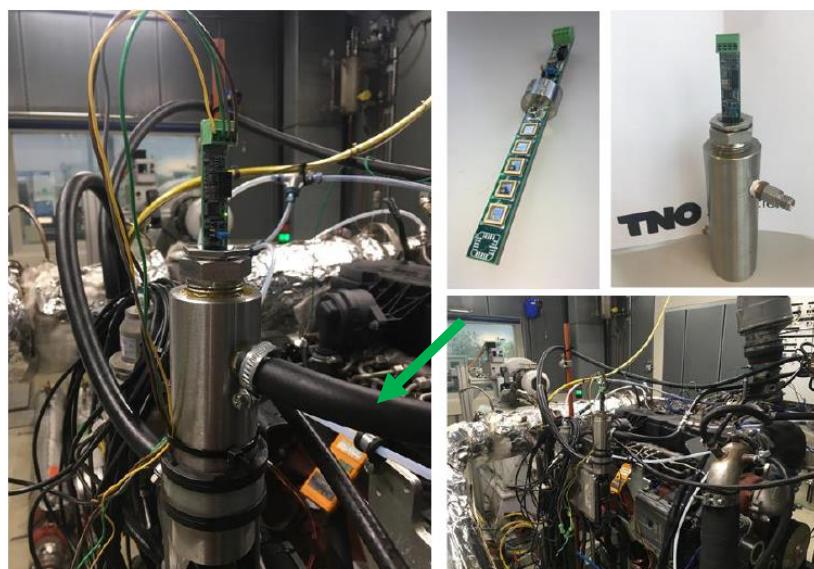


Figure 21: Installation of the ECC sensor in the fuel line of the TNO-powertrains dual fuel engine (top). Green arrow indicates location of the sensor. ECC MN measurement results (red) versus time and MN set point (black) computed by the NPL MN algorithm from known fuel (G20, MIX9) composition are displayed in the bottom. Pictures and results from [22].

More results on the MN sensors are described in publications of the project [24], [25].

The ECC sensor was further assessed for its applicability to detect methane slip (MS) and it was shown that the LNG ECC sensor is able to detect low concentrations of methane in nitrogen at low part per million (ppm) levels. Further it was noted that both the ECC and the FTIR sensors match the requirements of a methane sensitivity between 100-1000 ppm [22].

Within the project, actual MN's were determined from engine tests using the TUBS test facility shown in Figure 22. These are referred to as service MN's and the results were already used for the x-axis values in Figure 19 (left) and Figure 20. A single cylinder four-stroke spark ignition research engine was used for the engine tests. The MN of the gas mixtures is experimentally determined from its knocking behaviour and can then be

contrasted with the computed MN as shown in Figure 23. The reference gas mixtures of the project were used, which have a known composition. The regression line is very close to the diagonal  $y = x$  line. This implies that the service MN is not only qualitatively but also quantitatively comparable to the computed MN in this case [26]. In many cases, the service MN shows the same trend as the MN, however at different slope.

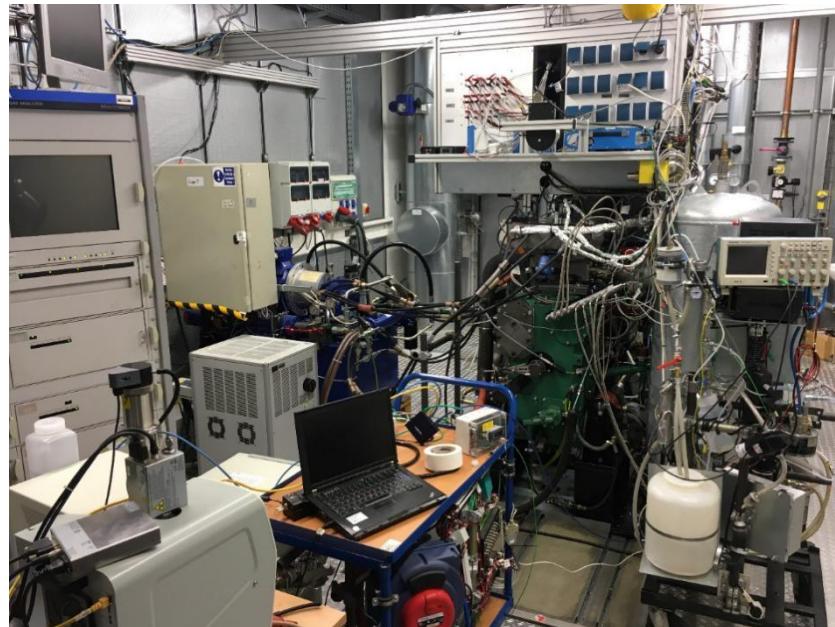


Figure 22: Engine set-up for service MN determination of the reference gases at TUBS. Picture taken from [26].

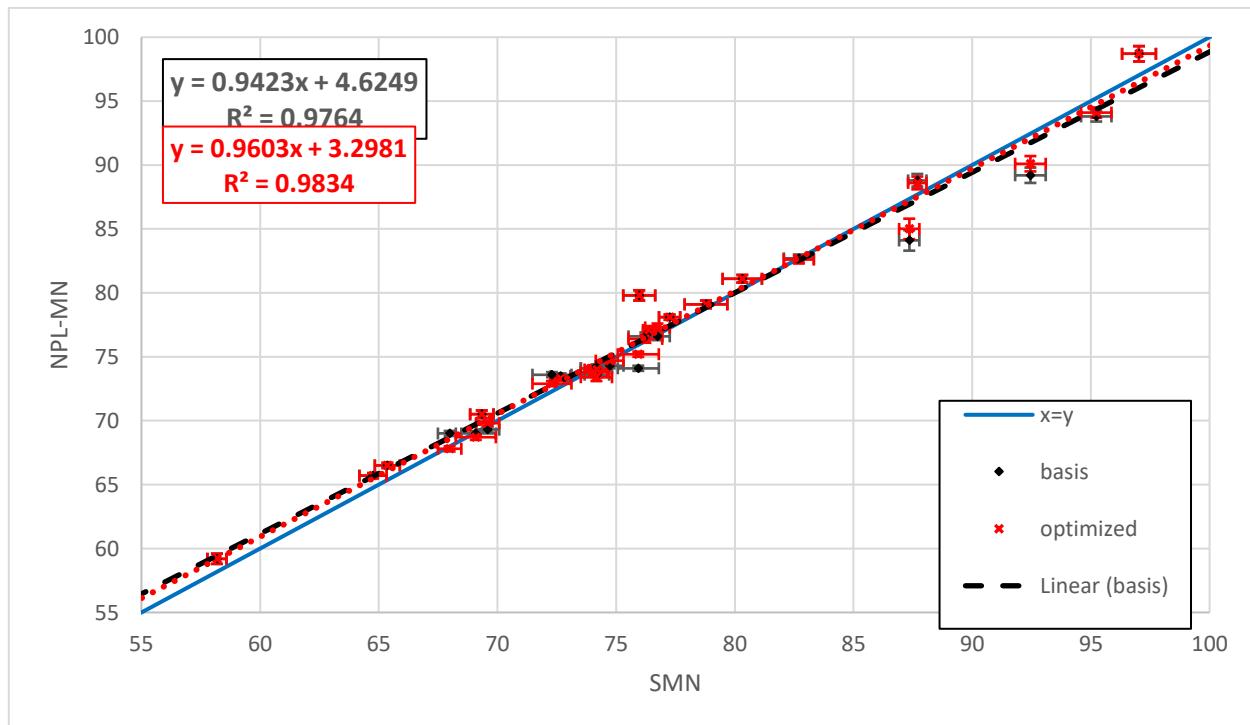


Figure 23: MN computed from composition using the NPL MN algorithm versus measured service methane number in TUBS engine tests showing the improvement in MN prediction from the optimized NPL MN algorithm developed in the project in red. Results taken from [26].

Further experiments were performed to determine the Ignition Delay Times (IDTs) of the project's reference gases for the purpose of improving existing MN algorithms for LNG engine management. Figure 24 shows two experimental setups at PTB for the determination of IDT's: the High Pressure Shock Tube (HPST, top left) and the Rapid Compression Machine (RCM, bottom left) with illustrations (right) of typical pressure traces for the determination of the IDT. Figure 25 shows the correlation between the measured IDTs and the reference gas MN's within a temperature range of 950 K and 1430 K [26].

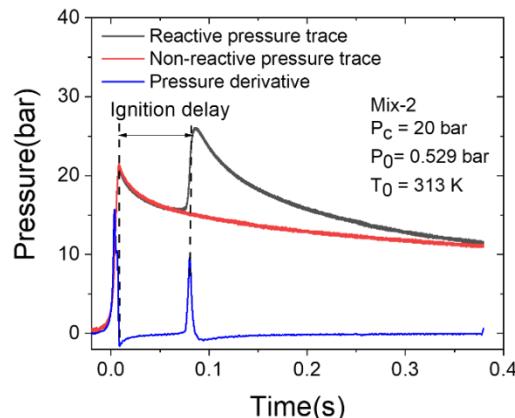
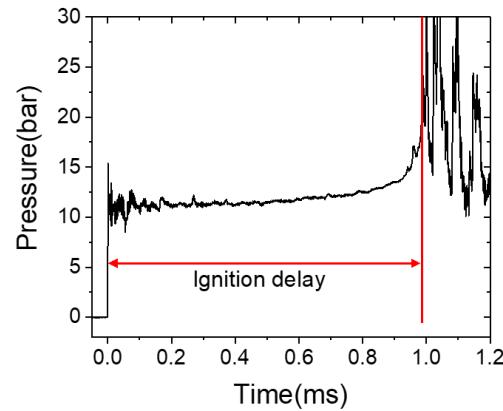


Figure 24: Experimental setups at PTB to determine the Ignition Delay Time (IDT) for reference gases with different MN's. High pressure shock tube (top left), typical pressure trace of shock tube measurements and the determination of the ignition delay (top right), rapid compression machine (bottom left), and typical pressure trace of RCM measurements and the determination of the ignition delay (bottom right). Pictures and graphs taken from [26] & [27].

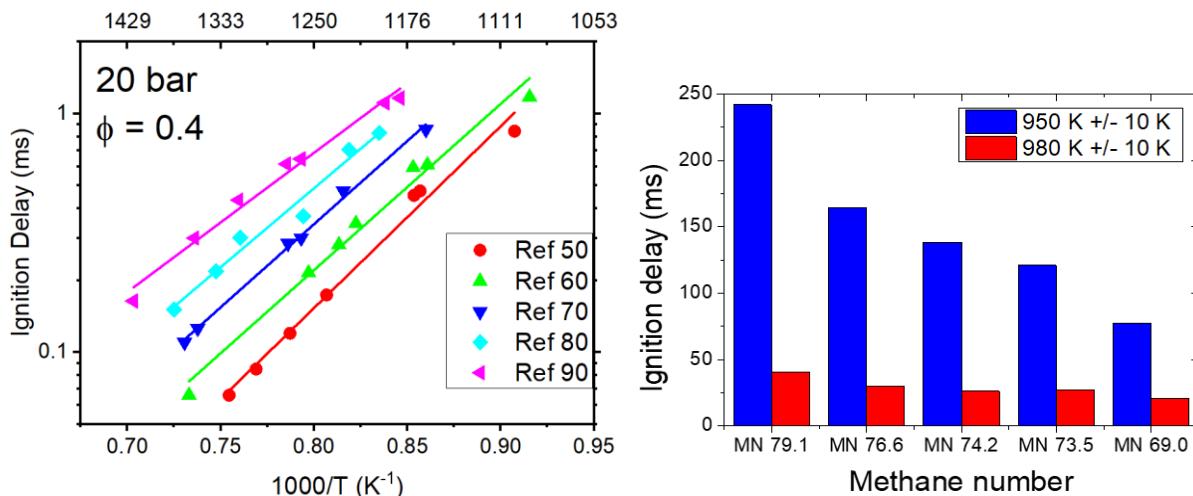


Figure 25: Measured ignition delay time (left) of the reference mixtures at 20 bar,  $\phi = 0.4$ , and temperature in the range of 950 K and 1430 K;  $\phi$  denotes the fuel-air equivalence ratio. Legend numbers indicate MN's; and correlation between IDTs and MNs at temperatures in the range of 950 K and 980 K at 20 bar, and  $\phi = 0.4$ . Graph taken from [26] which is published in [27].

Chemical kinetic modeling that can accurately predict the combustion and ignition behaviour of LNG with different MN's was performed and validated with the IDT-determination measurements. This was combined with the service MN measurements to develop an improved MN algorithm, in terms of increased accuracy (see red graph in Figure 23), that reflects real LNG engine behaviour. More can be read in [27] and [28].

Finally, within the project a good practice guide for the weighing of particulate filters used in LNG and LBG refuelling stations highlighting many of the issues and pitfalls associated with setting up traceable measurement, from on-site sampling to the final mass value determination for particulates in LNG/LBG, was published on the project website [29]. The guide describes a method for the accurate measurement of the collected particulate mass on filters by building on the expertise gained from the area of ambient air monitoring. Further to this it has offered guidance on the determination of particulate losses within sampling systems used at LNG refuelling stations.

In summary, the key outputs relating to the objective are:

- Validation of prototype LNG density and speed-of-sound measurement device.
- Three sensors to measure the composition and methane number were developed: (i) Coated Capacitive Chip (ECC), (ii) Tuneable Filter Infrared (TFIR), and (iii) Fourier Transform Infrared (FTIR)
- ECC, TFIR, and FTIR are able to measure the methane number with a 1.5 methane number unit accuracy with respect to the experimentally determined values.
- TFIR is a faster technique. The ECC sensor can be deployed in a larger (temperature and) pressure range. The FTIR sensor and correlation method is a reliable and robust laboratory technique for gas analysis. All analysers can be deployed as fuel quality monitors for LNG engine management.
- ECC sensor application was assessed for the detection of methane slip. Results indicate that application of a gas quality sensor for conventional dual fuel engines enables new optimisation strategies.
- A chemical kinetic model was developed that can accurately predict the combustion and ignition behavior of LNGs with different MNs.
- A SMN was determined for each LNG reference gas used in the project and the SMNs were compared to the MNs computed from different MN algorithms.
- A gravimetric method for the weighing of particulate filters used in LNG and LBG refuelling stations was developed and a good practice guide highlighting many of the issues and pitfalls associated with setting up traceable measurement in LNG/LBG, was published on the project website.

The project has clearly improved methods and developed (in-line) sensors for LNG density and cost-effective measurement of the gas composition, MN and MS meeting the research objective.

## 5 Impact

The project outcomes were disseminated through various channels. Project members have contributed to standard development, published journal papers, and presented at scientific and industrial conferences using the project's results (for further details see below).

### *Impact on industrial and other user communities*

The LNG research and calibration facility is commercially available for traceable calibration of LNG flow and composition instruments. This calibration facility supports the reliable LNG measurements needed for a variety of small and mid-scale LNG applications.

The advisory board was (re)established (a continuation from that in EMRP ENG60) and a new chair was elected. The advisory board was expanded to 35 members to include nine additional stakeholders from the engine manufacturer industry. The project board actively participated in the project's progress meetings, LNG metrology workshops and trainings. The project also disseminated the project's outcomes to industry working groups such as the GIIGNL custody transfer handbook working group and through the dissemination of technical reports on the project website.

The development of improved and traceable LNG flow, density, and composition measurements will support LNG custody transfer measurement traceability and stimulate the uptake of LNG as a transport fuel as the metrological infrastructure for quantity (flow and density) and quality (composition) is provided.

State-of-the-art equations of state (EOS) for LNG such as the GERG-2008 EOS were implemented in the Trend 4.0 software developed by RUB, and the .dll is made freely available for software development by interested parties involved in the computation of reliable thermodynamic properties from composition measurements.

The ECC sensor tested by the project was tested in the gas distribution network, where it successfully measured the composition of natural gas and biogas. This has led to interest from the company Bronkhorst High-Tech to further commercialise this sensor.

The TFIR sensor is further developed in a commercialisation project in conjunction with an engine company and a company specialised in optical measurement systems for online monitoring and control of a gas engine.

Further to this, the project has developed and improved a chemical-kinetic model which in reduced form can be used for LNG engine design and control. This model can be taken up by manufacturers of LNG ships and trucks.

### *Impact on the metrology and scientific communities*

In total, five journal papers stemming from the research were published, and another five are expected to be published once the peer review process is finalised. The project has achieved uptake by the metrological, industrial, standardisation, and scientific communities comprising the establishment of a calibration facility for LNG flow and composition, release of a new standard on dynamic LNG flow measurement (ISO 21903:2020), delivery of a cryogenic LDV standard, adoption of FTIR MN determination in a pre-operational plan of an LNG terminal, release of TREND software for thermodynamic modelling of LNG, and commercialisation projects of the ECC MN and TFIR MN sensors.

Three LNG metrology training meetings took place during the course of the project, one in Delft, The Netherlands alongside the project's kick-off meeting (August 2017), one in Aberdeen, UK, alongside the project's 18 month progress meeting (October 2018), and one as a teleconference alongside the project's 36 month progress meeting (May 2020). Stakeholders, members of the consortium, and a collaborator participated (maximum attendance was at approximately 50 attendees). Most presentations were

disseminated on the project website. The project has also disseminated its outcomes in numerous conferences, in scientific journals, and at LNG metrology workshops. Examples are: Oil and Gas Focus Group Meeting, 2018, United Kingdom, Kuwait 3rd Flow Measurement Conference, 2017, Kuwait, Eurosensors, 2018, Austria, GasTec, 2018, Spain, German Thermodinamic colloquim, 2018, Germany, Twentieth Symposium on Thermophysical Properties, 2018, United States, Clean Fossil Fuels Seminar, 2017, United Kingdom, Emerson Global Users Exchange, 2018, Netherlands, 10th International Symposium on Fluid Flow Measurement, 2018, Mexico, Global conference & exhibition – Innovative solutions in flow measurement and control, 2017, India, 7th International Metrology Conference – CAFMET 2018, 2018, India, XXIVth Encontro Luso Galego de Química, 2018, Portugal, 3. Tagung der Fuels Joint Research Group, 2018, Germany, 37th International Symposium on Combustion, 2018, Ireland, 117th General Assembly of the German Bunsen Society for Physical Chemistry, 2018, Germany, 6th International TMFB Conference, 2018, Germany, SAE World Congress, 2018, United States, Flomeko 2019, 2019, Portugal, Micro Fluidic Handling Systems, 2019, Netherlands, European Combustion Meeting, 2019, Portugal, 29. Deutscher Flammentag, 2019, Germany, 38th North Sea Flow Measurement Workshop, 2020, United Kingdom, Public Seminar at Technical University Dortmund, 2018, Germany.

The project is contributing to the calibration services know-how by studying the effects of upstream flow disturbances under ambient and cryogenic settings. Further to this, the cryogenic LDV flow sensor was validated in pertinent LNG custody transfer settings, providing a potential alternative to Coriolis and ultrasonic flow sensors for measuring flow. The collaboration with a liquefier manufacturer (EffecTech) contributed to the required metrological knowledge to validate composition measurements by means of a reference liquefier.

The cryogenic density meter was tested and validated with the aim to improve the measurement accuracy of LNG density measurements. Development is ongoing in terms of making one of the sensors suitable for use in an industrial setting.

The ECC and TFIR are currently being commercialised with applications of gas composition measurement in the gas grid and gas engine management by MN determination. Last, improved MN determination algorithms were developed as part of the project.

#### *Impact on relevant standards*

The knowledge and experience for LNG measurement and calibration were shared within various ISO-standard working groups, OIML technical committees and user groups such as the GIIGNL including: ISO/TC 28 WG20, ISO/TC28 SC4 and SC5, OIML/TC8 SC3 and SC6, ISO TC 193 WG8, the Gas Processors Association and American Petroleum Institute, and the GIIGNL Custody Transfer Handbook (CTH) working group.

Project partners NEL and VSL have developed, together with stakeholders representing instrument manufacturers and LNG end-users, a new standard “Refrigerated hydrocarbon fluids — Dynamic measurement — Requirements and guidelines for the calibration and installation of flowmeters used for liquefied natural gas (LNG) and other refrigerated hydrocarbon fluids” within the ISO/TC 28 WG20, chaired by VSL. The standard was published in February 2020. The release of the standard addresses key issues in the proper installation and calibration of flow meters used for dynamic measurement of LNG, which can help to prove this methodology in custody transfer applications supporting the use of LNG as a transitional fuel toward clean energy.

Project knowledge was also disseminated to the GIIGNL CTH working group. Cesame's cryogenic LDV standard was presented to the GIIGNL task force in September 2019 and is expected to be included as new route to traceability in the new revision of the GIIGNL CTH. NEL created a report on the outcomes of all LNG metrology projects which will be communicated with the CTH working group after project completion. Naturgy and Reganosa have communicated their advances on the FTIR sensor method for direct MN determination. Full adoption of LNG custody transfer measurements into the GIIGNL CTH will require methods to be fully supported by published datasets.

PTB, TUBS, NPL and VSL, disseminated the novel algorithm to calculate the MN from the LNG composition into the ISO/TC 193 WG8.

In collaboration with NIST, RUB provided input to the Gas Processors Association and American Petroleum Institute on the Enhanced Revised Klosek McKinley method for density calculations of liquefied natural gas (LNG).

VSL and NEL communicated the project outcomes within the NC 310 327 (Dutch mirror committee of CEN/TC282), the OIML/TC8 SC3 and SC8, ISO/TC28/SC4, and the ISO/TC28 SC5.

#### *Longer-term economic, social and environmental impacts*

LNG fuelled truck engines produce around 25 % less carbon dioxide ( $\text{CO}_2$ ) compared to diesel engines and 85 % less  $\text{NO}_x$ . Furthermore, they produce less noise and thus are the preferred option for deliveries in urban areas and city centres, especially in the early morning or late at night (when avoiding peak traffic). The developed metrological infrastructure directly supports the European Union's strategy for liquefied natural gas and gas storage.

The uptake of LNG and LBG as transport fuel will be underpinned by robust calibration services and more efficiently running engines. This in turn enables the uptake of the relatively clean LNG and even cleaner (bio-)LNG/LBG with concomitant economic and environmental benefits. Thereby making a significant contribution to the European "Clean Transport Fuel Strategy" which aims to reduce the emission of greenhouse gases, nitrogen oxides ( $\text{NO}_x$ ), sulphur dioxide ( $\text{SO}_2$ ), and particles.

The project has lasting impact by establishing metrological infrastructure for LNG flow, composition, and density, and the development of three types of cost-effective MN sensors for LNG engine management. Notable application examples are: i) the LNG research and calibration facility for flow and composition instruments, ii) improved calibration services know-how on cryogenic upstream flow disturbances and installation guidelines included into the ISO/DIS 21903:2018(E), iii) cryogenic LDV standard for flow measurement, iv) TREND 4.0 software for thermodynamic modelling of LNG, v) required metrological knowledge to validate composition measurements by means of a reference natural gas liquefier, v) The ECC and FTIR sensors are currently being applied for gas composition measurement in the gas grid and gas engine management by MN determination, vi) adoption of FTIR MN determination in a pre-operational plan of an LNG terminal, and vii) an improved MN algorithm. These project outcomes will support further developments for even more efficient LNG engine management, and for establishing LNG calibration traceability for the for small- and mid-scale applications and beyond.

## 6 List of publications

- I. Cavuoto, G., Lago, S., Giuliano Albo, P.A., Serazio, D., 2019, Speed of sound measurements in liquid methane ( $\text{CH}_4$ ) at cryogenic temperatures between (130 and 162) K and at pressures up to 10 MPa, *J. Chem. Thermodynamics*, 142, 106007. <https://doi.org/10.1016/j.jct.2019.106007>
- II. Shu, B., Vallabhuni, S.K., Zheng, J., Agarwal, S., Fernandes, R.X., 2020, Experimental and Modeling Studies on the Correlation Between Auto-Ignition Delays and the Methane Number of Liquefied Natural Gas (LNG) and Liquefied Biogas (LBG), *Frontiers in Mechanical Engineering*, 6, Article 47. <https://doi.org/10.3389/fmech.2020.00047>
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