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### INFLUENCE OF TECHNOLOGICAL PROCESSES ON THE UNCERTAINTY OF SYSTEMS MEASURING LIQUEFIED NATURAL GAS ENERGY

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**Abstract.** The article shows a new measuring technology and a statistical approach to control the measured values and parameters of technological processes that reduce the uncertainty of systems measuring the amount of liquefied natural gas energy (LNG). An increase in the consumption of liquefied natural gas requires the use of measuring technologies that take into account changes in physical and chemical properties, process parameters, and other factors affecting the uncertainty of measuring systems at subzero temperatures of working fluids. A measuring technology is presented that takes into account the influence of technological processes and changes in the properties of LNG. The application of a statistical approach to the control of measured values makes it possible to reduce the influence of technology and changes in physico-chemical properties on the uncertainty of the of systems measuring mass, volume, density and energy of LNG. The developed system of metrological support increases the accuracy of measurement systems of LNG energy by 1.5–2 times. The solution is relevant for information and measurement systems for determining the amount of energy of liquefied hydrogen.

**Keywords:** measurement system, uncertainty, energy, stratification, reliability, statistical, liquefied natural gas

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## ВЛИЯНИЕ ТЕХНОЛОГИЧЕСКИХ ПРОЦЕССОВ НА НЕОПРЕДЕЛЕННОСТЬ ИЗМЕРИТЕЛЬНЫХ СИСТЕМ КОЛИЧЕСТВА ЭНЕРГИИ СЖИЖЕННОГО ПРИРОДНОГО ГАЗА

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**Аннотация.** В статье показана новая измерительная технология и статистический подход контроля измеряемых величин и параметров технологических процессов, которые позволяют уменьшить неопределенность измерительных систем количества энергии сжиженного природного газа. Увеличение потребления сжиженного природного газа требует применения измерительных технологий, которые учитывают изменение физико-химических свойств, параметров технологического процесса, и других факторов, влияющих на неопределенность измерительных систем при отрицательных температурах рабочих жидкостей. Представлена измерительная технология, учитывающая влияние технологических процессов и изменение свойств СПГ. Применение статистического подхода при контроле измеряемых величин позволяет уменьшить влияние технологии и изменения физико-химических свойств рабочих углеводородных жидкостей на неопределенность ИИС массы, объема, плотности и энергии СПГ. Разработанная система метрологического обеспечения в 1,5–2 раза повышает точность измерительных систем энергии сжиженного природного газа. Решение актуально для информационно-измерительных систем определения количества энергии сжиженного водорода.

**Ключевые слова:** измерительная система, неопределенность, энергия, стратификация, достоверность, статистический, сжиженный природный газ

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### Introduction

Reception-transmission of liquefied natural gas (LNG) during transportation by sea vessels, reception and shipment to storage tanks are made on the basis of the amount of energy equivalent to the transferred volume of LNG. Measurements of the actual values of the volume of transferred LNG are currently carried out by a static method using level measurement systems when LNG is poured into a storage tank or tanker tanks using calibration and correction tables of tank capacity. The component composition and physico-chemical properties of LNG are determined in the laboratory from a sample selected by a special sampling device according to a given technological regulation [1, 5]. The values of density and calorific value are calculated based on the results of determining the component composition in laboratory conditions by gas chromatographic method [5, 13]. The methods for determining the density and calorific value when performing commodity transport operations (CTO) are described in the international standards GIIGNL [1–6, 8]. The accounting unit used in the calculations between the Seller and the Buyer is the amount of energy transferred: the equivalent of volume (mass) of the transferred LNG. LNG energy is defined in MJ or British thermal units MMBtu,  $1 \text{ MMBtu} = 1.055 \cdot 10^9 \text{ J}$  [5, 11].

The accuracy of measuring the level and the volume of LNG in the tank is determined by the uncertainty of the liquid–gas interface associated with the boiling point of LNG, which characterizes the

equilibrium state of any liquefied gas at boiling point. The uncertainty of level measurements depends on the uncertainty of tank calibration, on the technical condition, volume and type of tanks, sea waves, the type of system used and the thoroughness of measurements, the processes of “evaporation” and “boiling”, changes in the component composition of LNG [12]. The uncertainty of the tank calibration depends on the chosen calibration method and the discreteness in the construction of the calibration table [5, 11, 18, 19, 23–25].

When determining the actual volume in the tank, it is necessary to take into account the physico-chemical properties of a particular grade of LNG, for which the coefficients of volumetric expansion and compressibility differ. Additional influencing factors in determining the volume of LNG in the tank are the technical condition, dimensions, type of tank and thermal insulation, the magnitude of the coefficients of linear and volumetric expansion of materials and structural elements of tanks. During the voyage, it is important for the preservation of physicochemical properties to maintain the pressure in the tanker tanks greater than the pressure of LNG in shore tanks, this is necessary to limit evaporation during transportation, storage and unloading [11].

The daily volume of LNG evaporation in a tanker depends on the size of the tanks, the ratio of the liquid phase surface in the tanks to the loading volume, climatic and marine conditions (water temperature, air, sea condition, etc.) and thermodynamic characteristics of the loaded LNG (supercooling depth, etc.). Numerical values of evaporation vary from 0.18% to 0.25% of the total volume of LNG in existing tankers, and up to 0.10% for LNG tankers of new designs equipped with a new thermal insulation system [11, 28].

The static method of volume measurements is not applicable in the joint use of LNG storage terminals, when LNG belonging to several owners is pumped through the cargo fleet, loading ramps and supply pipelines: in this case, it is impossible to reconcile volumes with tankers by sellers [28]. In such cases, a dynamic method of measuring mass or volume by flow converters is used. The static methods employed today, with the use of level measurement systems in the tanker, make it possible to determine the volume of LNG with an uncertainty of 0.7–0.9% [28]. Factors affecting the accuracy of volume measurements in tanks using level systems are due to the heterogeneity/uncertainty of the measured liquid, the formation of a gas phase in the field of level measurements, which differs for different grades of LNG. These factors increase the uncertainty of determining the volume by  $\geq 1\%$  [11, 28]. Methods used in information and measurement systems (IMS) and sampling systems, methods for determining density, component composition and calorific value [16]. Volume measurements using level systems do not always allow determining the volume and amount of LNG energy with the required accuracy stated by the current regulatory and technical documents (RTDs), which is a natural change in the physico-chemical properties of LNG [11, 16, 26, 27].

The problem of determining the amount of LNG energy and transferring units of quantities from standards to measuring systems at subzero temperatures is caused by the difficulty in ensuring the homogeneous state of LNG during sampling and measurements. The increase in LNG consumption requires the use of advanced information and measurement systems to determine the amount of energy of liquefied natural gas (IMSLNG), including using dynamic systems with flow converters, appropriate measurement technologies that take into account changes in physical and chemical properties, process parameters, and other factors affecting the accuracy of determining the amount of energy. The developed IMSLNG should take into account these changes, and the applied measurement technologies of testing and control should ensure reliable measurements [11].

#### **Analysis of technological processes affecting the uncertainty of density and calorific value**

Determination of the density and the highest calorific value of the transmitted LNG is based on the determination of the component composition by gas chromatographic analysis. The method allows us to determine the volume fraction of the highest calorific value, and mass units are used for commercial operations, therefore it is necessary to measure the density of LNG [1–6, 8, 11, 15]. The contribution of the

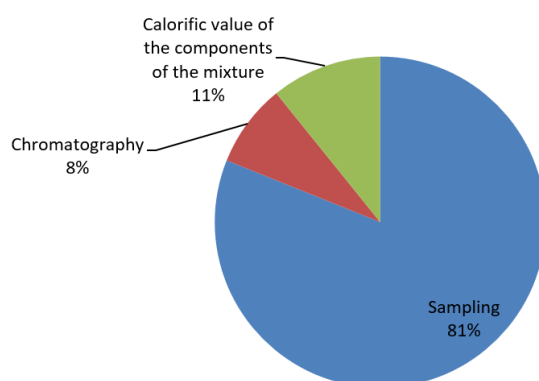


Fig. 1. Uncertainty budget of the highest calorific value

highest calorific value is up to 30% in the total uncertainty of the amount of transferred LNG energy. In the uncertainty of the highest calorific value, the greatest contribution is associated with sampling technology and can reach 81% (Fig. 1).

Density is the main input value in the equations for determining the mass and the highest calorific value of LNG. The contribution from the uncertainty of density is at least 40% in the uncertainty budget of the amount of transferred LNG energy.

GIIGNL standards allow two methods for determining LNG density [4, 5].

The first, a direct measurement method, using a density converter or a density channel of a mass flow converter installed in a tank or measuring pipeline. The second, indirect, currently used, consists in determining the density by component composition using gas chromatographic method. The main disadvantage of the first method is the lack of primary and working density standards that allow us to determine and confirm the actual density values in working conditions. When calibrating density converters and density channels of mass flow converters, liquids are currently used as a working medium, which differ significantly in their properties from the physico-chemical properties of LNG.

In the absolute majority of cases, water or petroleum products with a density and viscosity that differ from the properties of LNG are used for calibration, when conversion coefficients are used to bring them to negative temperatures. Recalculation of calibration results performed at positive temperatures of 20–25 °C to the operating conditions of LNG with a temperature of minus 162.5 °C and a pressure of 0.5–1.0 MPa introduces additional uncertainty, the value of which can reach 1.0 kg/m<sup>3</sup> or more. Therefore, direct density measurement methods currently have limited application and are used only for technological control [12]. The disadvantages of the second, indirect, measurement method include significant contributions from the sum of uncertainties determined by the sampling location, the type of sampling system selected, the sampling and storage method, the thoroughness with which the operator took the sample, the type of analyzer and the measurement method [8, 9, 11, 19, 20].

The main problem of LNG density measurements for direct and indirect methods is the difficulty of ensuring a homogeneous state of the liquid flow in the area of the converter and sampling system. In fact, the flow is inhomogeneous, consisting of a liquid and a gas phase, in which the number and ratio of phases vary. Moreover, in some cases, measurements become impossible, or the measurement results exceed the permissible limits of uncertainty, and this is due to the technological process, instability of the composition and properties of LNG [8, 9, 13, 16, 29].

The actual values of uncertainties in the results of LNG density measurements by density converters or density channels of mass converters caused by the above conditions are at least 3–5 kg/m<sup>3</sup> [11]. In laboratory conditions, the Kleinram and Wagner hydrostatic method or the Klosek-McKinley pycnometric method can be used. These methods are currently not used in commercial operations due to the length of

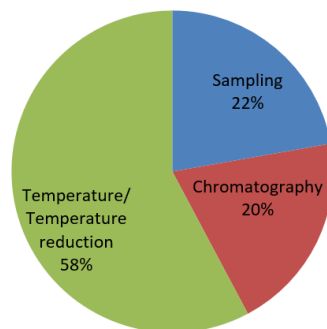


Fig. 2. Uncertainty budget of density measurements

the measurement process and the high cost of unique equipment, but are used only for research in laboratory conditions [5, 8, 9].

The density measured in the laboratory from the selected sample will always differs from the density of LNG in the measuring pipeline or in the tank. The problem of density measurements at the site of operation remains unresolved [20]. A similar situation was with measurements of the density of commercial oil during commercial operations, when in the mid-eighties of the last century, the urgent task was to find the reasons for exceeding the error in the mass of oil at CTO. The reason for exceeding the permissible uncertainty in mass was the measurement of oil density by hydrometers in the laboratory, when light fractions evaporated, and the hydrometer measured oil with a different density. Moreover, with an increase in the time interval between sampling and measurements, the density of oil also changed more. The reason for the excess of uncertainty in the mass of oil was eliminated after special reference density measuring instruments that receive a unit of magnitude from state standards appeared in Russia. Density standards are designed to monitor metrological characteristics and verify density converters at the place of operation [8, 9, 11].

The second group of indirect methods, which are the main ones at present, includes various methods for determining the density by the component composition of LNG [11], for example: the Watson method, the Elfacuten method, the Miller graphical method, the Heese method, the Klosek-McKinley method. A promising method of spectroscopy proposed by C.V. Raman is based on molecular level studies, a method that is less dependent on changes in pressure and flow velocity in the sampling system line and with better repeatability of measurement results. However, the method has limited application due to the duration of measurements, high cost and qualification of personnel [5, 11].

In accordance with the requirements of the GIIGNL standard [5], the method of determining density by component composition and temperature makes the greatest contribution to the uncertainty of determining density. Uncertainty of component composition determination by gas chromatography is 0.09% according to GIIGNL [4, 5, 13]. The values of contributions to the total standard uncertainty of calorific value measured by indirect, gas chromatographic measurement method are presented in the diagrams in Fig. 1 and Fig. 2. The greatest contribution to the uncertainty budgets of density measurements and higher calorific value is made by sampling, temperature measurement and temperature reduction.

The analysis of density and calorific value uncertainties proves the necessity of using a direct method of LNG density measurements, the use of which eliminates the largest components associated with evaporation and temperature conversions increasing the accuracy of density and calorific value measurements. [11, 23–25].

The actual values of the uncertainty of determining the component composition are much higher, depend on factors not previously taken into account, and this is the uncertainty of sampling, which depends

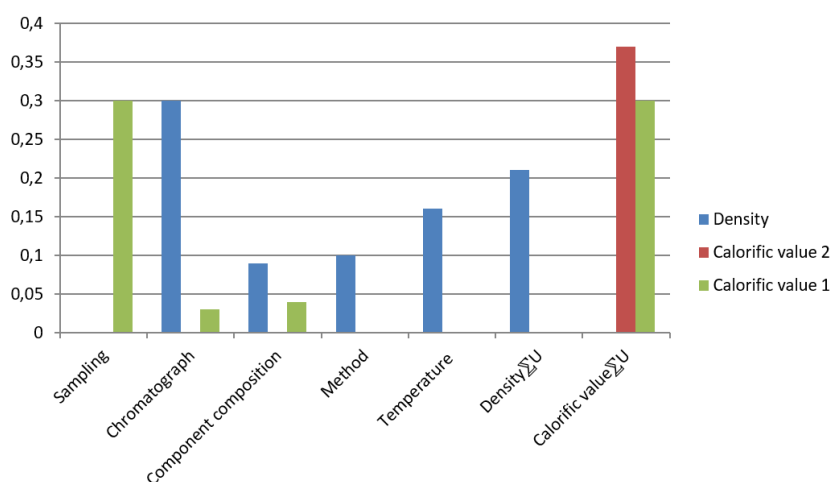


Fig. 3. Uncertainty budget of calorific value of IM system

on the phase state of the flow [10]. Density and calorific value in the total uncertainty of the amount of LNG energy are presented in Fig. 3.

Taking into account the real values of uncertainties, we obtain the LNG density determined by the GIIGNL methods [5], reduced by temperature and pressure to operating conditions, which gives results significantly different from the actual values of  $\geq 10 \text{ kg/m}^3$ .

#### **Analysis of the influence of technological processes on the stratification of flows of multicomponent liquids**

The main influencing factors on stratification and homogenization of a multicomponent LNG stream are velocity, pressure of the flow in the pipeline and pressure of saturated vapors [24]. The results of studies of the flow processes of mixtures of hydrocarbon liquids have shown that stratification of the LNG mixture flow occurs at speeds of less than 1.5–2 m/s and pressures of less than 0.4 MPa, the values of which may vary depending on the component composition of a particular LNG grade [11].

Complete mixing of the flow occurs at speeds of more than 2.5–3 m/s and pipeline pressure of at least 0.5–0.7 MPa as shown in Fig. 4.

When the flow velocity exceeds more than 5–6 m/s, the flow becomes stratified again, further increase in pressure fails to return the flow to a homogeneous state.

To reduce the total standard uncertainty of LNG energy, it is necessary to reduce the uncertainties associated with volume measurement and sampling. It is necessary to apply methods to increase the representativeness of sampling and to assess the reliability of sampling, density measurement and to develop equations of natural gas state taking into account the influencing factors [13, 16, 29].

#### **Analysis of the impact of technological processes on the reliability of sampling**

During the period of pumping a batch of LNG with the help of a control multipoint (tubular) probe, profile tests are carried out, during which the distribution of the estimated LNG quality indicators is determined, for example the value of the density or methane content selected along the pipeline section [13]. As a base value, the sampling results obtained from the control probe, the number of sampling points along the pipeline section ( $n+2$ ), compared with the working probe are taken [13]. When removing the profile from each point located along the diameter of the pipeline, each sample is taken into a separate container. All selected samples are analyzed, and based on the obtained measurement results, a graphical dependence of the distribution of the values of these values over the diameter of the pipeline is constructed. According to the obtained dependence, the averaged values of the value for the period corresponding to sampling for this flow profile are determined [13]:

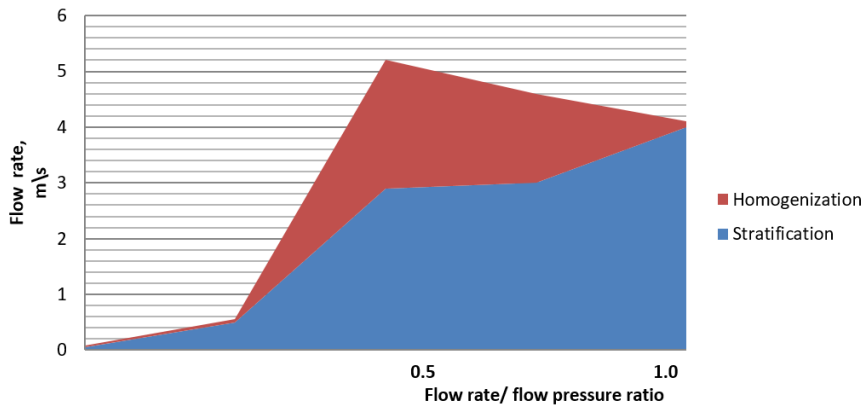


Fig. 4. Phase state of the LNG mixture flow depending on the velocity and pressure

$$W_{avg_j} = \frac{1}{D} \sum_{i=1}^n W_i \Delta X_i, \quad (1)$$

where  $D$  is pipeline internal diameter, mm;  $\Delta X_i$  is distance between sampling points, mm;  $W_i$  is quality indicator value of  $i^{\text{th}}$  point sample, %;  $n$  is number of sampling points [13].

During the period of LNG pumping, at least three profiles are removed at the operating flow rate. The value of the quality indicator is estimated for the period of the set time of sampling of the test sample during shipment/loading. The basic value of the quality indicator value for the period of sampling of the combined sample under study is calculated by the formula:

$$W_{bas} = \frac{1}{T} \sum_{j=1}^k W_{avg_j} \Delta t_j, \quad (2)$$

where  $\Delta t_j$  is time period for which  $j^{\text{th}}$  profile is defined, minutes;  $W_{avg_j}$  is average value of a quantity in  $j^{\text{th}}$  profile by equation (6), %;  $T$  is time of sampling of the combined sample under research, minutes [13].

The assessment of the reliability of sampling is considered satisfactory when the deviations of the average values of the estimated value according to the results of measurements of three flow profiles do not exceed 5% [13].

The results of studies on the deviation of the density and content of methane in the LNG mixture were carried out on a pipeline with a diameter of 250 mm at five cross-section points. Figure 5 shows density deviations of more than 5% (417–442 kg/m<sup>3</sup>) along the pipeline section of stratified flow and homogeneous LNG flow, when density deviations along the pipeline section do not exceed 2% (430–438 kg/m<sup>3</sup>). To create a homogeneous flow, the pressure in the pipeline after the sampling point was increased by 24% [13].

Figure 6 shows deviations of methane content of more than 7% (88–95%) along the pipeline section for stratified flow and homogeneous LNG flow, when density deviations along the pipeline section do not exceed 4.5% (92–96%).

Estimation of sampling uncertainty. The research results have shown that sampling depends on the phase state of the working media flow, which makes the greatest contribution to the uncertainty budget of the measured values: component composition, density and calorific value – the main indicators that require verification during LNG reception and transmission operations. With the experimental evaluation method, a measurement procedure is performed to directly assess the uncertainty of the measurement result. With the theoretical evaluation method, each source of uncertainty is quantitatively assessed separately and in combination in the budget in accordance with the methodology adopted for this purpose

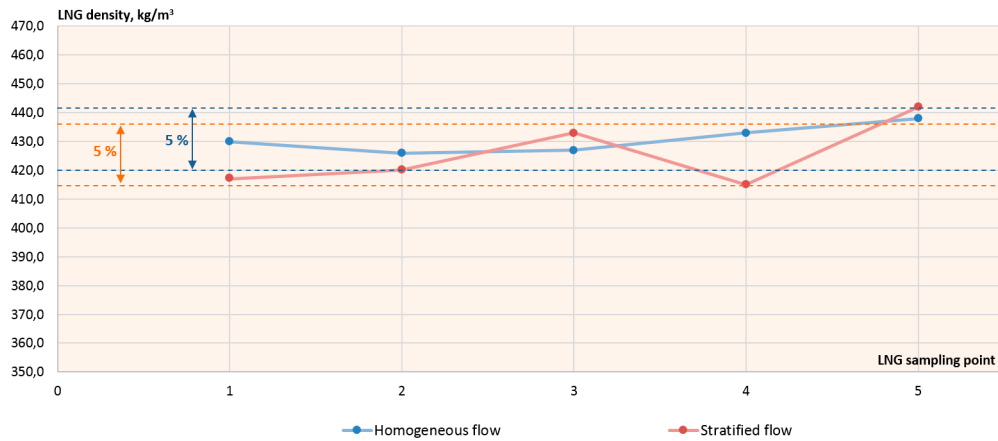


Fig. 5. Sampling reliability estimate in density determination

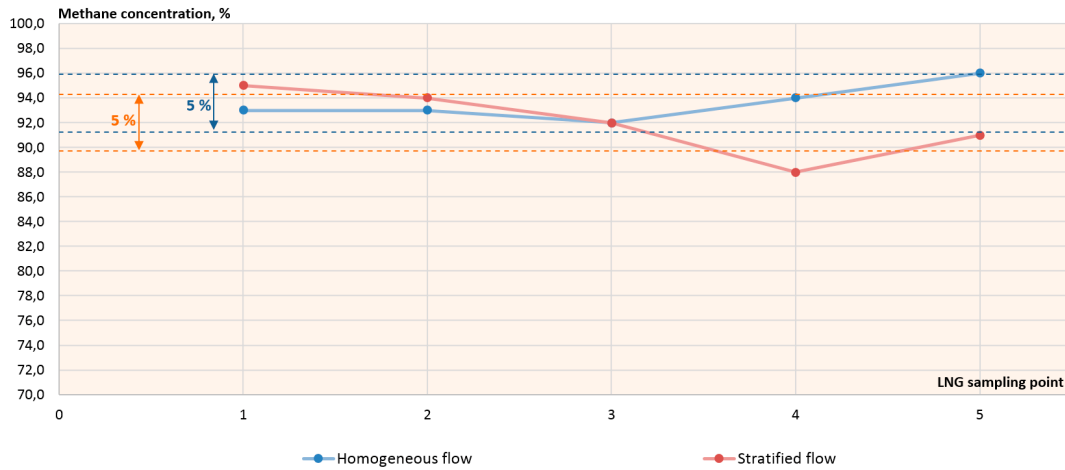


Fig. 6. Sampling reliability estimate in methane content determination

for assessing the reliability of sampling [16]. The dispersed nature of the LNG mixture leads to a dispersion of the measurement results of the samples taken, which is caused by the different composition of the samples actually taken causing an additional contribution to the total uncertainty [16]. The experimental approach is widely used in practice, for example, the “double sampling method”. The implementation of the method requires less time and costs, which is especially important for one-time studies of various target objects. The “double sampling method” usually involves the work of one sampling operator and one sampling scheme; the same plan can be used with different operators to take into account the subjective contribution to uncertainty. The random effects model of a single sampling object can be described by the following formula:

$$X = X_{true} + \varepsilon_{sample} + \varepsilon_{analysis}, \quad (3)$$

where  $X_{true}$  is actual value of the measured variable;  $\varepsilon_{sample}$  is uncertainty component due to with drawal procedure;  $\varepsilon_{analysis}$  is uncertainty component due to selected analysis procedure.

For a single target object, when the sources of variance are independent, the variance of the measurement result  $\sigma^2$  can be represented by the equation:



$$\begin{aligned}
 u_c(V) = & \left[ u_c(V_{cal})^2 + u_c(V_{phase\ boundary})^2 + u_c(K_{temp})^2 + \right. \\
 & \left. + u_c(K_{cal\ op\ temp})^2 + u_c(V_{cool})^2 + u_c(V_{heat})^2 \right]^{1/2}, \quad (4) \\
 \sigma_{meas}^2 = & \sigma_{sample}^2 + \sigma_{analysis}^2,
 \end{aligned}$$

when approximating the variance by statistical methods, we obtain:

$$\mathbf{u}_{meas}^2 = \mathbf{u}_{sample}^2 + \mathbf{u}_{analysis}^2, \quad (5)$$

To improve reliability, it is suggested to consider sampling uncertainty  $\mathbf{u}_{sample}$  with the following components not included before:  $\mathbf{u}_{flow}$  as uncertainty due to heterogeneity of LNG multi-component flow and  $\mathbf{u}_{sample\ type}$  as uncertainty due to probe type:

$$\mathbf{u}_{sample}^2 = \mathbf{u}_{flow}^2 + \mathbf{u}_{sample\ type}^2, \quad (6)$$

where  $\mathbf{u}_{flow}$  is uncertainty due to natural stratification and low density component distribution in the upper section of pipeline and higher density components in the lower section. Currently, only the uncertainty of the analysis and evaporation of the selected sample during preparation is evaluated. The presence of a gas phase in the sampling zone must be monitored using the phase state indicator (PSI) of the flow. It is also permissible to control the flow at the sampling site using a pressure converter installed after the sampling probe to control the back pressure value recommended for specific technological conditions and the LNG grade [16]. In this case, the sampling uncertainty is determined taking into account the contribution of sampling variance not previously taken into account: the phase state of the flow and the type of sampling system [16]. Then the sampling uncertainty is calculated using a formula that takes into account the possible stratification of the multicomponent flow, the type of sampling probe used, the uncertainty of analysis/measurement in determining the component composition and the uncertainty associated with sample storage and sample preparation for measurements [28, 29]:

$$\mathbf{u}_{sampling} = \left( \mathbf{u}_{flow}^2 + \mathbf{u}_{sample\ time}^2 + \mathbf{u}_{analysis}^2 + \mathbf{u}_{sample\ prep.}^2 \right)^{1/2}, \quad (7)$$

#### Analysis of the impact of technological processes on the uncertainty of calculating the amount of LNG energy

To increase the efficiency and reduce the uncertainty of IMSLNG in determining the amount of LNG energy, it is recommended to apply an improved static method for determining volume using currently used level measurement systems, currently used and a new, dynamic method for measuring mass, volume and density using volume, mass and density converters, a direct method of static measurements. The formula for calculating the total amount of energy  $E$ , MJ, enclosed in the volume of transferred LNG at CTO is proposed by the GIIGNL standard [5].

$$E = V_{LNG} \cdot D_{LNG} \cdot GCV_{LNG} - E_{transf. gas} \pm E_{fuel gas}, \quad (8)$$

where  $V_{LNG}$  is volume,  $m^3$ ;  $D_{LNG}$  is density,  $kg/m^3$ ;  $GCV_{LNG}$  is gross calorific value, MJ/kg;  $E_{transf. gas}$  is energy of transferred gas (gaseous phase), MJ;  $E_{fuel gas}$  is energy of auxiliary gas per period of loading, MJ, positive when loading and negative when offloading [29].

To measure the actual values of the transferred LNG energy, four equations of state were developed for liquefied natural gas. The equations can be used for IMSLNG, in static method of measurement, which is

based on indirect measurements of liquefied natural gas volume with level measurement system according to current GIIGNL standard [5].

They can be applied to dynamic IMSLNG using indirect and direct methods of volume and mass measurements, volume and mass converters, and for a new method of direct static measurements of the mass of liquefied natural gas.

**Improvement of IMSLNG using an indirect method of static volume measurements using level measurement systems and laboratory analyzers of density and component composition.** To determine the amount of energy, the following formula is proposed, which takes into account the influencing factors of technological processes related to the determination of density, volume and calorific value, not previously taken into account [29].

$$E = V_{LNG} \cdot D_{LNG} \cdot GCV_{LNG} - E_{transf. gas} \pm E_{fuel gas} \pm D_{LNG} \cdot GCV_{LNG} \cdot u_c(V) \pm \pm V_{LNG} \cdot GCV_{LNG} \cdot u_c(D_{LNG}) \pm V_{LNG} \cdot D_{LNG} \cdot u_c(GCV_{LNG}), \quad (9)$$

where  $V_{LNG}$  is volume of loaded/offloaded LNG, m<sup>3</sup>;  $D_{LNG}$  is density of LNG mix, kg/m<sup>3</sup>;  $GCV_{LNG}$  is gross calorific value of LNG, MJ/kg. It is necessary to consider additional values such as standard measurement uncertainty of volume, density, and calorific value.

When assessing the uncertainty of determining the volume, Eq. (10), in addition to the GIIGNL standard [5], we need to take into account the uncertainties of technological processes affecting the result of measurements of the LNG level of the liquid-gas interface, temperature differences during calibration and operation of the tank, gradients of temperature changes of the tank material during loading and unloading.

$$u_c(V) = \left[ u_c(V_{cal})^2 + u_c(V_{phase boundary})^2 + u_c(K_{temp})^2 + u_c(K_{cal op temp})^2 + u_c(V_{cool})^2 + u_c(V_{heat})^2 \right]^{1/2}, \quad (10)$$

where  $u_c(V_{cal})$  is combined standard volume uncertainty, which depends on tank calibration curve, level measurement in liquid-gas interface boundary  $u_c(V_{phase boundary})$ , uncertainty of temperature measurement along tank height  $u_c(K_{temp})$ , uncertainty of temperature difference while tank calibration and service  $u_c(K_{cal op temp})$  taking into account temperature gradient during tank walls cooling/heating  $u_c(V_{cool})$  and  $u_c(V_{heat})$  while loading/offloading respectively (in m<sup>3</sup>).

When density uncertainty is estimated by gas chromatography and using Eq. (11), there are additionally included uncertainties due to chromatograph type and method of defining the composition, tank temperature difference while sampling and in laboratory  $u_c(K_{temp})$ , flow phase condition in sampling area, and calorific value [18].

$$u_c(D) = \left[ u_c(D_t)^2 + u_c(D_p)^2 + u_c(D_{sampling})^2 + u_c(D_{chrom})^2 + u_c(D_{composition})^2 + u_c(K_{temp})^2 + u_c(D_{phase})^2 + u_c(GCV_{sampling})^2 \right]^{1/2}, \quad (11)$$

where  $u_c(D)$  is combined standard uncertainty of density, depending on temperature  $u_c(D_t)$ , pressure  $u_c(D_p)$ , sampling and sample storage  $u_c(D_{sampling})$ , density measurement by composition  $u_c(D_{chrom})$ , composition measurement  $u_c(D_{comp})$ , and heterogeneity of LNG mix in sampling area  $u_c(D_{phase})$ , kg/m<sup>3</sup>. In case of measurement with density meter,  $u_c(D)$  is sum of uncertainty due to meter calibration  $u_c(D_{cal})$  and uncertainty due to LNG phase condition.

$$u_c(D) = \left[ u_c(D_{cal})^2 + u_c(D_{phase})^2 \right]^{1/2}, \quad (12)$$

Using the direct method of density measurement allows us to eliminate uncertainties due to sampling and sample storage, reducing by temperature and pressure, composition determination, and chromatograph type, which contribute up to 40% to the total uncertainty.

When calorific value uncertainty is estimated using Eq. (13), there are additionally included uncertainties due to determination of composition, density, flow phase condition, and tank temperature difference in sampling area and in laboratory  $u_c(K_{temp})$ .

$$u_c(GCV_{LNG}) = \left[ u_c(GCV_{LNG\ component})^2 + u_c(GCV_{sampling})^2 + u_c(D)^2 + u_c(GCV_{LNG\ phase})^2 + u_c(K_{temp})^2 \right]^{1/2}, \quad (13)$$

where  $u_c(GCV_{LNG})$  is combined standard uncertainty of gas calorific value, depending on gas composition  $u_c(GCV_{LNG\ component})$ , sampling and sample storage  $u_c(GCV_{LNG\ sampling})$ , heterogeneity of LNG mix in sampling area  $u_c(GCV_{LNG\ phase})$ , MJ/kg [18].

**Improvement of IMSLNG using an indirect method of dynamic measurements with and volume flow transducers and in-line density transducers.** In order to determine energy content, we propose the following formula which additionally includes the factors related with determination of density, volume, and calorific value not included before [14]:

$$E = V_{LNG} \cdot D_{LNG} \cdot GCV_{LNG} - E_{transf. gas} \pm E_{fuel gas} \pm D_{LNG} \cdot GCV_{LNG} \cdot u'_c(V) \pm \pm V_{LNG} \cdot GCV_{LNG} \cdot u_c(D_{LNG}) \pm V_{LNG} \cdot D_{LNG} \cdot u_c(GCV_{LNG}), \quad (14)$$

where  $u'_c(V)$  is combined standard uncertainty of volume measurement, depending on volume flow meter calibration uncertainty  $u_c(V_{cal m})$ , presence of gaseous phase in liquid  $u_c(V_{phase})$ , operating liquid temperature difference during calibration and loading/offloading  $u_c(V_{temp})$ , m<sup>3</sup>.

When using Eq. (15) to estimate volume uncertainty, there are additionally included uncertainties due to flow phase condition and meter temperature difference during calibration and operation  $u_c(K_{temp})$ .

$$u_c(V) = \left[ u_c(V_{cal m})^2 + u_c(V_{phase})^2 + u_c(V_{temp})^2 \right]^{1/2}, \quad (15)$$

$$u_c(D) = \left[ u_c(D_t)^2 + u_c(D_p)^2 + u_c(D_{sampling})^2 + u_c(D_{chrome})^2 + u_c(D_{composition})^2 + u_c(K_{temp})^2 + u_c(D_{phase})^2 + u_c(CGV_{sampling})^2 \right]^{1/2}, \quad (16)$$

where  $u_c(D)$  is combined standard uncertainty of density, depending on temperature  $u_c(D_t)$ , pressure  $u_c(D_p)$  in liquid and gas phases, sampling and sample storage  $u_c(D_{sampling})$ , density measurement by composition  $u_c(D_{chromat})$ , composition measurement  $u_c(D_{composition})$ , phase condition of LNG mix in sampling area  $u_c(D_{phase})$ , kg/m<sup>3</sup>.

$$u_c(GCV_{LNG}) = \left[ u_c(GCV_{LNG\ comp})^2 + u_c(GCV_{LNG\ sampling})^2 \right]^{1/2} \quad (17)$$

$$+ u_c \left( GCV_{LNG\ phase} \right)^2 + u_c (D)^2 + u_c \left( K_{temp} \right)^2 \Big]^{1/2}, \quad (17)$$

where  $u_c(GCV_{LNG})$  is combined standard uncertainty of gas calorific value determination, depending on gas composition  $u_c(GCV_{LNG\ comp})$ , sampling and sample storage  $u_c(GCV_{LNG\ sampling})$ , heterogeneity of LNG mix in sampling area  $u_c(GCV_{LNG\ phase})$ , MJ/kg.

**Improvement of IMSLNG using a direct method of dynamic measurements with mass and mass flow measuring transducers.** To determine the amount of energy, the following formula is proposed, which takes into account factors related to the measurement of mass and calorific value. The amount of LNG energy is determined by a formula that takes into account [29]:

$$E = M_{LNG} \cdot GCV_{LNG} - E_{transf. gas} \pm E_{fuel gas} \pm GCV_{LNG} \cdot u_c(M) \pm M_{LNG} \cdot u_c(GCV_{LNG}), \quad (18)$$

$$u_c(M) = \left[ u_c(M_{cal m})^2 + u_c(M_{phase})^2 + u_c(M_{temp})^2 \right]^{1/2}, \quad (19)$$

where  $u_c(M)$  is combined standard uncertainty of mass measurement, depending on mass flow meter calibration uncertainty  $u_c(V_{cal m})$ , presence of gaseous phase in liquid  $u_c(M_{phase})$ , operating liquid temperature difference while calibration and loading/offloading  $u_c(M_{temp})$ , kg.

When using the Eq. (20) to estimate calorific value uncertainty, there are additionally included uncertainties due to effect of sampling and sample storage, LNG flow phase condition, and temperature difference in laboratory and in field  $u_c(K_{temp})$ ,

$$u_c(GCV_{LNG}) = \left[ u_c(GCV_{LNG\ comp})^2 + u_c(GCV_{LNG\ sampling})^2 + u_c(GCV_{LNG\ phase})^2 + u_c(D)^2 + u_c(K_{temp})^2 \right]^{1/2}, \quad (20)$$

where  $u_c(GCV_{LNG})$  is combined standard uncertainty of gas calorific value measurement, depending on gas composition  $u_c(GCV_{LNG\ comp})$ , sampling and sample storage  $u_c(GCV_{LNG\ sampling})$ , phase condition of LNG mix in sampling area  $u_c(GCV_{LNG\ phase})$ , MJ/kg [29].

**Improvement of IMSLNG using the developed direct method of static measurements of LNG mass [29]** in a tank with the use of weight measuring systems. The amount of energy is determined by the formula:

$$E = M'_{LNG} (\Delta\phi_{LNG}) \cdot K_{mass} \cdot GCV_{LNG} - E_{transf. gas} \pm E_{fuel gas} \pm M_{LNG} (\Delta\phi_{LNG}) \cdot u_c(GCV_{LNG}) \pm GCV_{LNG} \cdot u_c(M_{LNG}), \quad (21)$$

where  $M'_{LNG}$  is LNG mass measured in tank, kg;  $\Delta\phi_{LNG}$  is correction coefficient, taking into account the acceleration of gravity and deviation of the geographical area of service from the calibration site for load cells, nondimensional quantity;  $u_c(M_{LNG})$  is weight measuring system calibration uncertainty, kg.

$$u_c(GCV_{LNG}) = \left[ u_c(GCV_{LNG\ comp})^2 + u_c(GCV_{LNG\ sampling})^2 + u_c(GCV_{LNG\ phase})^2 + u_c(D)^2 \right]^{1/2}, \quad (22)$$

where  $u_c(GCV_{LNG})$  is combined standard uncertainty of gas calorific value determination, depending on gas composition  $u_c(GCV_{LNG\ comp})$ , sampling and sample storage  $u_c(GCV_{LNG\ sampling})$ , phase condition of LNG mix in sampling area  $u_c(GCV_{LNG\ phase})$ , MJ/kg.

Correction coefficient  $K_{mass}$  for effect of buoyancy in the air when measuring LNG mass in tank by gravimetric method:

$$K_{mass} = 1 - \frac{D_{air}}{D_{LNG}}, \quad (23)$$

where  $D_{air}$  is ambient air density, kg/m<sup>3</sup>;  $D_{LNG}$  is liquefied natural gas density, liquid, kg/m<sup>3</sup>.

**Analysis of the control system/diagnostics of technological processes and the impact on the IMS of the amount of LNG energy**

The continuous monitoring system, based on the results of the evaluation of the measured value and the parameters of the technological process, develops solutions that allow performing two measurement tasks.

**The first measuring task of the control system.** Collection, processing and analysis of measuring information coming from level, temperature, pressure, gas chromatographs converters and systems, and if available, from mass, volume and density flow converters. Output of information about technological parameters: “normal state” and “change of technological process parameters”, requiring operator intervention. In case of a violation of the phase state of LNG (the presence of gas), which affects the uncertainty of measurements of mass, volume, level, density and component composition, recommendations are issued for changing the parameters of the technological process or for applying correction coefficients to determine the actual amount of energy transmitted by LNG. IMSLNG measurement results are monitored during loading, unloading, transportation and storage. The list of controlled parameters is determined by the technological regulations and measurement methodology.

**The second measuring task of the control system.** Monitoring of IMSLNG characteristics, measuring transducers and systems of level, temperature, pressure and gas chromatographs, dynamic density, volume and density converters. Collection and analysis of values of technological process parameters. The frequency of monitoring of IMSLNG characteristics is specified in the technological regulations and measurement methodology. The application of a statistical approach to the assessment of measured values and parameters of technological processes, using the improved method of control charts by W. Shuhart [17, 21, 29] allows us to ensure and maintain processes at a stable level to perform reliable measurements, Table 1.

Table 1

**Determination of the limits of permissible deviations of IMSLNG (level, volume, density and calorific value)**

Measured (estimated) value	Standard values of the measured value are not used		Standard values of the measured value are set by the measurement procedure	
	Estimated value	2–4σ control limits	Estimated value	2–4σ control limits
$p$	$\bar{p}$	$\bar{p} \pm 3\sqrt{\frac{\bar{p}(1-\bar{p})}{n}}$	$p_0$	$p_0 \pm 3\sqrt{\frac{p_0(1-p_0)}{n}}$
$np$	$n\bar{p}$	$n\bar{p} \pm 3\sqrt{n\bar{p}(1-\bar{p})}$	$np_0$	$np_0 \pm 3\sqrt{np_0(1-p_0)}$
$c$	$\bar{c}$	$\bar{c} \pm 3\sqrt{\bar{c}}$	$c_0$	$c_0 \pm 3\sqrt{c_0}$
$u$	$\bar{u}$	$\bar{u} \pm 3\sqrt{\frac{\bar{u}}{n}}$	$u_0$	$u_0 \pm 3\sqrt{\frac{u_0}{n}}$

Note:  $n$  is the volume of the subgroup;  $p$  is the proportion of nonconformities in the subgroup;  $np$  is the number of inconsistencies in the subgroup;  $c$  is the number of subgroup mismatches;  $u$  is the number of inconsistencies per unit in the subgroup;  $\sigma$  is the standard deviation of the process of the estimated value.

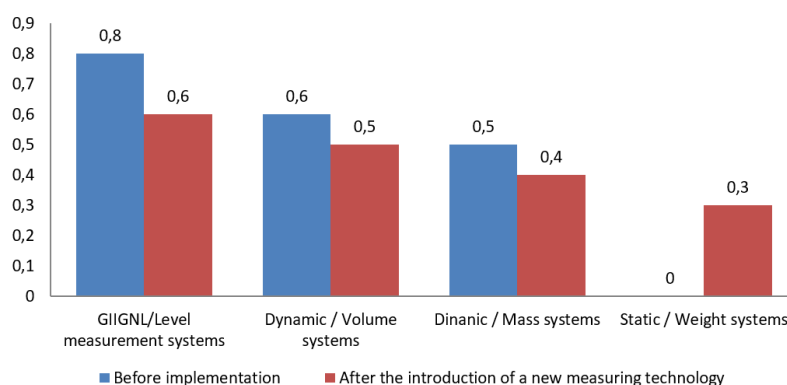


Fig. 7. Comparison of IMSLNG uncertainty budgets

The use of measurement technologies aimed at reducing the uncertainty of IMSLNG using the developed methods for assessing the reliability of sampling, taking into account changes in technological processes and monitoring / diagnostic systems do not involve large capital expenditures that can be attributed to maintenance and operation, or work carried out as part of technical re-equipment. Reducing the uncertainty of IMSLNG makes it possible to reduce maintenance and operation costs by 1.5–2 times and determine the amount of transferred LNG, excluding losses associated with changes in technological processes and physico-chemical properties. But the most accurate method, independent of the parameters of technological processes, in determining the amount of LNG energy, Figure 7, is the direct mass method using weight measuring systems [28].

IMS based on weight measurement systems allows you to determine the amount of energy with an expanded uncertainty of not more than 0.3%. In this case, four factors remain the main sources of uncertainty – a direct measurement of mass and density, a sampling system and an indirect gas chromatographic method for determining the calorific value [28].

### Conclusion

The inclusion of factors influencing the determination of the amount of LNG energy into the uncertainty budget does not increase the uncertainty budget, but allows to reduce uncertainty by eliminating and reducing the influencing factors due to the process technology. Therefore this provides the expansion of the functionality of the IMS and conditions for determining the actual values of the component composition, density, calorific value, volume and mass of LNG. Based on the equations of state of LNG technological processes, a mathematical model was developed for calculating the amount of transferred energy, which makes it possible to reduce uncertainties by ensuring control of the phase state of the flow and reliable sampling. It allows selecting the parameters of technological processes to evaluate the results of determining the volume, mass, component composition, density, calorific value, taking into account their contribution to the budget of uncertainties in the amount of LNG energy [29].

The developed methods and new measuring technology, the use of a statistical approach in the control of measured quantities and parameters of technological processes reduce the influence of technology and physical and chemical properties on the uncertainty of the IMS of mass, volume, density and energy of LNG [14]. This solution develops the system of metrological support of IMS using static and dynamic methods, reduces operating costs and financial risks in the course of receiving and transferring LNG goods and transport operations in the Russian Federation and export operations. The metrological support system makes it possible to increase the accuracy of the IMS of LNG energy by 1.5–2 times. The solution is relevant for IMS to determine the amount of energy of liquefied hydrogen.

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