Investigation of measurement uncertainties in LNG density and energy for custody transfer

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Abstract

In the custody transfer of fuels such as Liquefied Natural Gas (LNG), energy measurement accuracy is critical. The important elements in calculating transferred energy are the volume, density and the superior calorific value (SCV). In this paper we have used a new approach to carefully study the various uncertainty factors contributing to two of these elements: the density and SCV. For density, six methods were evaluated, namely the method of extended corresponding states (ECS), Hiza method, revised Klosek-McKinley method (RKM), Groupe Européen de Recherche Gazière method (GERG-2004), GERG-2008 method, and EOS-LNG method, to estimate the modeling error in density calculation. Other factors affecting LNG density estimation (LNG composition, reference densimeter, temperature and pressure) have also been investigated to derive the total uncertainty in LNG density estimation. For SCV, the uncertainty has been analysed with consideration of uncertainties in the composition and the SCV of LNG components. The expanded uncertainty in LNG energy during custody transfer is estimated to be 0.58% (k=2) after considering the uncertainties due to Membrane-type tank's unloaded LNG volume, SCV, density and their correlation effect.

Keywords: LNG metrology, LNG density calculation, LNG custody transfer, LNG energy, Measurement uncertainty, LNG superior calorific value, LNG composition, Tank volume uncertainty

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Nomenclature

u_{model}	standard uncertainty in LNG density due to modeling error
$D_{c,i}$	calculated densities of the LNG sample *i*
$D_{m,i}$	measured densities of the LNG sample *i*
AAD	averaged absolute deviation in calculated LNG density using multiple samples
e_m	maximum error in calculated LNG density
u_d	relative standard uncertainty of a densimeter
u_{sam}	relative standard uncertainty of a LNG sampler/vaporiser
u_{GC}	relative standard uncertainty for gas composition analysis
u_r	repeatability of a LNG sampler/vaporiser
u_{X_i}	relative standard uncertainty in measuring LNG composition
u_a	standard uncertainty in density due to sampling, vaporisation and analysis errors
e_t	maximum relative error in LNG density

u_T	relative standard uncertainty in LNG density due to temperature measurement
u_p	relative standard uncertainty in LNG density due to pressure measurement
u_ρ	relative standard uncertainty in the calculated LNG density
δ_M	mass of the transferred LNG
ρ	calculated LNG density
V	measured volume of the transferred LNG
T	LNG temperature
p	LNG pressure
SCV_{mass}	superior calorific value of natural gas in mass
E_d	energy of displaced gas
E_e	energy of gas consumed in ship's engine room
u_v	standard uncertainty in unloaded/loaded volume of a LNG tank
X_i	mole fraction of each component in LNG
SCV_i	*molar* SCV of each component in LNG
M_i	molecular weight of each component in LNG
u_{SCV}	standard uncertainty of LNG's SCV in mass
r_{12}	correlation coefficient between u_ρ and u_{SCV}
U_{energy}	expanded uncertainty of the transferred LNG energy

Acronym

SCV	Superior calorific value
LNG	Liquefied Natural Gas
ECS	Extended corresponding states
RKM	Revised Klosek-McKinley method
GERG	Groupe Européen de Recherche Gazière
EOS	Equation of state
GC	Gas chromatography
CMF	Coriolis mass flowmeter
GUM	Guide to the expression of Uncertainty in Measurement
ERKM	Enhanced revised Klosek-McKinley method

1. Introduction

The demand for Liquefied Natural Gas (LNG) is expected to double by 2040 to 700 million tonnes per annum [1]. Accurate measurement of transferred LNG energy with low uncertainty is required in LNG custody transfer [2]. LNG shipment values are often in the range of €30 - €40 million. A small error in the determination of the density and the superior calorific value (SCV) of the transferred LNG will have a significant financial impact on the exporter/importer. A 1% error in transferred energy equates to €300,000 - €400,000 in misallocation during LNG custody transfer. It is therefore important to understand the uncertainty associated with LNG energy transfer measurements and quantify it accurately. Previous work reported in [3] has focused on the sensitivity of the uncertainty in LNG energy transfer to LNG composition and temperature changes. In this paper, we focus on detailed uncertainty estimation in the LNG density, LNG SCV, composition analysis, temperature and pressure measurement effects.

We have considered a case where the LNG energy transferred is determined from LNG density, SCV and the measured volume. The estimation of uncertainty in LNG density and SCV has been described in [4] without detailed analysis. The LNG density was derived using the revised Klosek-McKinley (RKM) method, and its uncertainty was estimated using this specific method, with uncertainty contribution from the composition, modelling error (based on NBS publication 1080), calibration gas composition and temperature. Pressure, densimeter error and sampling factors are not considered in [4]. The SCV uncertainty values used in [4] are based on ISO 6976 [5], however, more recent development on improved measurement of SCV [6–10] can provide smaller uncertainties for measurement of SCV.

A new approach is presented in this paper to study the uncertainty in LNG density and SCV, with consideration of the latest development in SCV measurement, new equation of state for the thermodynamic properties of natural gases and most recent densimeter results for reference measurement of LNG density. Various uncertainty factors have been considered. We have derived the uncertainties in LNG SCV and density which are caused by the uncertainties in LNG composition. These are used to give a reliable estimate of the uncertainties in LNG density and SCV, which are then used to derive the uncertainty in transferred LNG energy. Throughout this paper, our expanded uncertainty estimation considers a coverage factor k=2 which corresponds to a 95% confidence level.

2. LNG density calculation

The density of LNG within an LNG tank can be calculated based on its compositions, temperature and pressure. There are a few methods to calculate LNG density, such as the method of extended corresponding states (ECS), the Hiza method, the RKM method, the Groupe Européen de Recherche Gazière wide-range Equation of State method (GERG-2004), GERG-2008 method and the most recent EOS-LNG method [11–17] .

Five LNG samples with methane composition ranging between 81% and 98% have been used to evaluate 4 calculation methods (ECS, Hiza, RKM, GERG-2004). The LNG sample compositions and their measured densities are obtained from publications of Haynes [18] and Orrit [19]. Table 1 shows the composition of the five LNG samples. Table 2 gives the experimental data and measured densities of 5 samples, from the open literature [18,19]. The LNG temperature range is from 112 K to 115 K, with pressure ranging from 0.12 to 0.22 MPa. The measured LNG densities range from 434.4 to 490.9 kg/m^3.

Table 3 shows the calculated densities of the LNG samples obtained using the ECS, Hiza, RKM and GERG-2004 methods, with comparison to the measured densities shown in Table 2. The software tools used to calculate the densities are: GasCalc 2.2 (for GERG-2004), Visual Basic &

Excel (for ECS) and Excel (for Hiza method and RKM). To evaluate the calculation error of each LNG density calculation method, the averaged absolute deviation (AAD) for the 5 LNG samples is obtained by

$$\mathrm{AAD} = \frac{\sum_{i=1}^{5}\left(|D_{c,i} - D_{m,i}\,|/D_{m,i}\right)}{5} \qquad (1)$$

where $D_{c,i}$ and $D_{m,i}$ are the calculated and measured densities of the LNG sample *i*. Table 3 shows that, among the four methods, the maximum *AAD* is 0.117% for density calculation using Hiza method. The *AAD* is 0.067% for the RKM method, which is commonly used in the field of LNG custody transfer.

Table 1: Composition of five LNG samples used for calculating their LNG density, from open literature [18,19] .

	Sample A	**Sample B**	**Sample C**	**Sample D**	**Sample E**
Component	**Mole Fraction (mol mol $^{-1}$)**				
Methane	0.81249	0.85341	0.90068	0.9513	0.9754
Ethane	0.08484	0.07898	0.06537	0.0189	0.0046
Propane	0.04931	0.04729	0.02200	0.0105	0.0053
Isobutane	0	0.00854	0.00291	0.0045	0.0049
n-Butane	0.02708	0.00992	0.00284	0.0042	0.0052
Isopentane	0	0.00097	0.00010	0	0
n-Pentane	0	0.00089	0.00011	0	0
n-Hexane	0	0	0	0	0
Nitrogen	0.02628	0	0.00599	0.0106	0.0046

Table 2: Experimental data and measured densities for the five LNG samples from open literature [18,19] (the temperature and pressure are used for calculating LNG density of the samples).

	Sample A	**Sample B**	**Sample C**	**Sample D**	**Sample E**
Molecular weight, g/mol	20.0706	19.3587	17.9026	17.0957	16.7363
Temperature, K	115	115	115	112.37	112.11
Pressure, MPa	0.2214	0.1172	0.1456	0.134	0.116
Measured densities, kg/m^3	490.8506	477.3197	454.0099	441.6	434.4
Expanded relative measurement uncertainty for LNG density	0.12%	0.12%	0.12%	0.028%	0.028%
Source of experimental data	Haynes, 1982, [18]	Haynes, 1982, [18]	Haynes, 1982, [18]	Orrit, 1983, [19]	Orrit, 1983, [19]
	Example g	Example m	Example q	Table 80	Table 79

Table 3: Comparison of the measured densities ([18,19]) and calculated densities using ECS, Hiza method, RKM and GERG-2004 for the five LNG samples shown in Table 1 and Table 2.

LNG sample	Measured density (kg/m^3)	Extended Corresponding States (ECS)		Hiza Method		Revised Klosek-McKinley (RKM)		GERG-2004	
		Calculated density (kg/m^3)	Deviation in calculated density (%)	Calculated Density (kg/m^3)	Deviation in calculated density (%)	Calculated Density (kg/m^3)	Deviation in calculated density (%)	Calculated Density (kg/m^3)	Deviation in calculated density (%)
A	490.851	490.940	0.018	489.671	-0.240	490.789	-0.013	490.010	-0.171
B	477.320	477.458	0.029	447.021	-0.062	477.315	-0.001	476.322	-0.209
C	454.010	453.942	-0.015	453.265	-0.164	453.756	-0.056	453.352	-0.145
D	441.600	442.138	0.122	441.681	0.018	442.111	0.116	441.644	0.010
E	434.400	435.066	0.153	434.833	0.100	435.050	0.150	434.592	0.044
***AAD* (%)**			**0.067**		**0.117**		**0.067**		**0.116**

In [20], five LNG samples with various compositions shown in Table 4 have been measured by a new LNG densimeter [21] with an expanded uncertainty of 0.044%. The measured densities have been compared to the calculated densities using the GERG-2008 method. The deviations of the calculated densities (using GERG-2008 method) from the measured values are 0.05%, 0.13%, 0.12%, 0.23% and 0.07%, respectively for the 5 samples shown in Table 4 [20] , so the maximum deviation in calculated density is 0.23% using GERG-2008 method.

A new calculation method (EOS-LNG) has been proposed to improve the accuracy of calculated LNG density in [16]. The maximum deviation of the measured and calculated densities using this new method is 0.05% [16] for the five LNG samples shown in Table 4.

In comparison with our analysis of the four methods (where the maximum deviation is 0.117%) and the GERG-2008 method, the new EOS-LNG method gives the smallest deviation from the measured density value. In section 3.1, we will use the EOS-LNG method to estimate the standard uncertainty due to modelling in LNG density calculation.

Table 4: Composition of LNG samples used for calculating their LNG densities via GERG-2008 and EOS-LNG method (as reported in [16,20]).

	LNG-Libya	LNG-Oman	LNG-2	LNG-5	LNG-7
Component	**Mole Fraction (mol mol $^{-1}$)**				
Methane	0.81562	0.87885	0.84636	0.87972	0.97890
Ethane	0.13374	0.07274	0.12800	0.07240	0.00999
Propane	0.03679	0.02926	0.01499	0.02900	0.00497
n-Butane	0.00688	0.01565	0.00209	0.00692	0.00209
Isobutane	0	0	0.00220	0.00643	0.00177
n-Pentane	0	0	0.00030	0.00101	0.00016
Isopentane	0	0	0.00020	0.00110	0.00018
Nitrogen	0.00695	0.00351	0.00585	0.00344	0.00193

3. Uncertainty in LNG density estimation

The uncertainties due to the LNG density calculation model (u_{model}), reference densimeter uncertainty (u_d), LNG sampling/vaporisation/composition (u_a), temperature measurement (u_T) and pressure measurement (u_p) will all contribute to the uncertainty in LNG density estimation. A detailed analysis is given in this section. The measurement uncertainties are determined according to the Guide to the expression of Uncertainty in Measurement (GUM) [22].

Though the EOS-LNG gives slightly smaller deviation than RKM in the calculated density, the differences in the error limits in density (caused by the uncertainty in composition and temperature) estimated using RKM or EOS-LNG method are negligible. The RKM method is easier to implement, so we will use this method to calculate the error limits in density caused by the uncertainties in composition and temperature.

3.1. Modeling error

The maximum error in the calculated densities was found to be e_m = 0.05% (based on EOS-LNG method), since we have selected the EOS-LNG method after evaluating the 6 calculation methods (ECS, Hiza, RKM, GERG-2004, GERG-2008 and EOS-LNG) in section 2. A few other methods give larger deviations, but these methods were not considered in the uncertainty analysis for modelling error. The most suitable method for density calculation in custody transfer is the EOS-LNG, which gives the smallest deviation from the measured density value.

The uncertainty in LNG density due to the calculation model error is estimated to be

$$u_{model} = \frac{e_m}{\sqrt{3}} = \frac{0.05\%}{\sqrt{3}} = 0.029\% \qquad (2)$$

where we have assumed a rectangular distribution for this modelling error [22].

3.2. Uncertainty of reference densimeter

A magnetic suspension densimeter was used to measure the density of LNG sample A, B and C in [18] to verify the accuracy of various density calculation methods. The expanded uncertainty for LNG density measurement using this magnetic suspension densimeter is estimated by Haynes to be 0.12% (k=2) in [18]. A cryodensimeter was used to measure the density of LNG samples D and E in [19], which has an expanded uncertainty of 0.028% (k=2) as reported by Orrit [19].

A single-sinker densimeter [21] was used to measure the density of the five LNG samples in [16,20]. This system used an analytical balance with magnetic suspension coupling and a Vapor-Liquid Equilibrium (VLE) cell for the density measurement. This measurement system has an expanded uncertainty of 0.044% (k=2), so its standard uncertainty is 0.022% , assuming a normal distribution [22].

We have selected EOS-LNG method to estimate the uncertainty for modelling error which has been verified by the single-sinker densimeter via multiple LNG samples. Thus, the relative standard uncertainty of the single-sinker densimeter (0.022%) [21] is included as one of the standard uncertainty components in evaluating the LNG density uncertainty: u_d =0.022% .

3.3. Uncertainty due to LNG sampling, vaporisation and composition analysis

LNG can be analysed directly in liquid phase using Raman spectroscopy technique [23]. The advantage is that LNG composition can be obtained by Raman spectroscopy directly without the vaporisation step. This can reduce the uncertainty in LNG composition which is caused by the sampler and vaporiser. Measurement of the mole fractions of individual components contained in a liquid stream of LNG is made by analysing the Raman spectra. The accuracy of Raman spectroscopy measurement is determined by cryogenic gravimetric standards of LNG mixtures with known compositions [24,25]. The expanded uncertainty for Raman spectroscopy in energy value measurement is about 0.14% after validation using an LNG composition reference standard in liquid phase [26]. The Raman spectroscopy's sensitivity needs to be improved to detect nitrogen and heavier hydrocarbons with concentrations below 200 ppm [23].

Currently the more established practice for sampling and analysing LNG is to use a liquid sampler and vaporiser, followed by gas chromatography (GC) analysis [4]. The LNG should be in a subcooled condition at the sampling point. The vaporised natural gas is analysed by a GC to yield the compositions of LNG components, which are used to calculate the LNG density. Some heat or pressure variation in the LNG sampler may produce LNG partial evaporation, due to big difference among the boiling points of LNG components. Partial vaporisation should be avoided as it will change the composition of collected LNG sample in a vaporiser. When there is no partial vaporisation before the vaporiser unit, the relative standard uncertainty of a well-designed sampler/vaporiser is u_{sam} = 0.10% with a repeatability of u_r = 0.05% (k=1) [27].

According to [28], the relative standard uncertainty of natural gas mixtures used to calibrate the GC is estimated to be 0.22% and the relative standard uncertainty for gas composition analysis is 0.20%. Thus the relative standard uncertainty for the mole fractions of natural gas mixture components is derived by combining these two relative uncertainties to give u_{GC} = $\sqrt{0.22^2 + 0.20^2}$ = 0.30%.

The relative standard uncertainty in measuring LNG composition (u_{X_i}) is derived by combining the relative standard uncertainties due to LNG sampler/vaporiser (u_{sam} = 0.10%), sampling repeatability (u_r = 0.05%) and composition analysis (u_{GC} = 0.30%), as given below

$$u_{X_i} = \sqrt{u_{sam}^2 + u_r^2 + u_{GC}^2} = 0.32\% \qquad (3)$$

We have studied the effect of composition measurement uncertainty (u_{X_i}) on the calculated LNG density using LNG sample B in Table 1 at 115 K. The calculated density for this LNG sample is 477.33 kg m^{-3}. The sample B has been chosen since it is a heavy mixture for LNG, which can be used to evaluate the uncertainty contribution from composition measurement as a "worst-case" scenario.

We have used the GUM approach [22] for the uncertainty propagation from composition measurement uncertainty of each LNG component (u_i) to the uncertainty of LNG density through the sensitivity coefficients C_i . The composition of each component has a relative standard uncertainty of 0.32%, thus the corresponding absolute uncertainty value (u_i) is derived by multiplying the composition (mol mol^{-1}) by 0.32%. The sensitivity coefficient C_i for each LNG component is derived using the RKM method [4]. Table 5 shows the combined standard uncertainty in calculated LNG density due to LNG composition measurement uncertainty is $\sqrt{\sum_i C_i^2\, u_i^2} = 0.16$ kg m^{-3}, thus the relative standard uncertainty in LNG density is u_a = 0.16/477.33 = 0.033% (k=1).

We have checked the uncertainties in LNG density due to composition measurement uncertainties at other temperature, which are similar to the uncertainty value derived at 115K. The LNG temperature change does not affect the combined uncertainty contribution due to composition measurement uncertainties (which is estimated to be 0.033% as shown in Table 5).

Table 5: Propagation of the standard uncertainties in composition of each component (u_i) to the standard uncertainty of the LNG's density (using the composition of LNG sample B in Table 1 with a calculated density of 477.33 kg m^{-3} at T=115K,)

Source of uncertainty component	**Uncertainty value u_i , (mol mol $^{-1}$)**	**Sensitivity coefficient C_i, (kg m-3) /(mol mol $^{-1}$)**	**Standard uncertainty in density, $C_i\, u_i$ (kg m^{-3})**
composition of methane	0.0027	48.94	0.13
composition of ethane	0.00025	207.75	0.053
composition of propane	0.00015	410.05	0.062
composition of n-butane	0.000032	616.51	0.020
composition of isobutane	0.000027	593.87	0.016
composition of n-pentane	0.0000031	768.89	0.0024
composition of isopentane	0.0000028	838.01	0.0024
Combined standard uncertainty in LNG density, u_ρ (kg/m $^{-3}$) **LNG density: 477.33 kg m^{-3}**			0.16
Relative standard uncertainty in LNG density (k=1)			0.033%

3.4. LNG temperature measurement uncertainty

The LNG temperature is measured by temperature sensors inside the LNG tank. Each temperature sensor has an expanded uncertainty of 0.5 K as reported in [4]. Thus, the maximum error in measured LNG temperature is assumed to be 0.5 K, which leads to a maximum relative error in the calculated LNG density, e_t = 0.144%, estimated using RKM method [4,12]. The relative standard uncertainty in LNG density due to temperature measurement error is estimated to be $u_T = e_t/\sqrt{3}$ = 0.083% (assuming a rectangular distribution [22]).

3.5. LNG pressure measurement uncertainty

The LNG pressure is up to 4 barg within a typical LNG carrier's pressure tank (type-C) [29]. The effect of pressure measurement error on the LNG density can be estimated using the Enhanced RKM (ERKM) method provided in [30]. The pressure sensor's accuracy for pressure measurement is within 1% of full range during custody transfer [31] , which is 0.25 bar for a typical pressure sensor range up to 25 bar. If we assume the maximum error in LNG pressure measurement is 0.25 bar, the maximum relative error in calculated LNG density due to pressure measurement error is estimated to be 0.007% using ERKM method (at T=135 K, p=5 bar). The standard uncertainty in LNG density due to LNG pressure measurement error is therefore estimated as u_p = 0.007%/√3 =0.004% (assuming a rectangular distribution).

3.6. Combined uncertainty in LNG density estimation

The relative uncertainty due to the modelling for LNG density calculation (u_{model}), reference densimeter measurement (u_d), LNG sampling/vaporisation/composition analysis (u_a), temperature measurement error (u_T) and pressure measurement error (u_p) will all contribute to the final uncertainty in LNG density estimation. Here we consider the case where the LNG is sampled, vaporised and analysed by a GC. The combined uncertainty in the calculated LNG density can be estimated as

$$u_\rho = \sqrt{u_{model}^2 + u_d^2 + u_a^2 + u_T^2 + u_p^2} \qquad (4)$$

The combined relative standard uncertainty for the calculated LNG density is estimated to be u_ρ = 0.097% (k=1) as shown in Table 6.

We have used the ratio $(u_i)^2/(u_\rho)^2$ to show the percentage of each uncertainty contribution factor (u_i) in Table 6. The dominating contribution is from the temperature measurement uncertainty, which has a percentage of 74.0%. The uncertainty in LNG density can be reduced if the LNG temperature can be measured with smaller uncertainty.

Table 6: Uncertainty budget to derive the relative standard uncertainty of the calculated LNG density

Uncertainty component	**Probability distribution**	**Relative standard uncertainty contribution, u_i (%)**	**Contribution percentage $(u_i)^2/(u_\rho)^2$**
Modeling for LNG density calculation, u_{model}	rectangular	0.029	8.9%
Uncertainty due to LNG sampling, vaporisation and composition analysis, u_a	normal	0.033	11.7%
Reference densimeter measurement uncertainty, u_d	normal	0.022	5.2%
Temperature measurement uncertainty (0.5 K), u_T	rectangular	0.083	74.0%
Pressure measurement uncertainty, u_p	rectangular	0.004	0.2%
Combined relative standard uncertainty of LNG density, u_ρ (k=1), %		0.097	

4. Uncertainty in LNG energy transfer

In LNG custody transfer operations, the transferred LNG energy from/to an LNG carrier can be derived as

$$E = \delta_M \times SCV - E_d \pm E_e = V \times \rho \times SCV - E_d \pm E_e \qquad (5)$$

where δ_M is the mass of the transferred LNG, $\delta_M = V \times \rho$, ρ is the calculated LNG density, V is the measured volume of the transferred LNG, and SCV is the superior calorific value of LNG in mass, E_d is the energy of the displaced gas (gas sent back to the LNG carrier or the shore tank during the unloading/loading processes) and E_e is the energy of the gas consumed in the ship's engine room. In this paper, we have considered the unloaded or loaded LNG volume as the measured volume of the transferred LNG.

It has been shown in [4] that the total uncertainty in estimated energy of displaced gas is 0.003% and the total uncertainty in engine-consumed gas is 0.0006%. Both uncertainties are negligible and can be ignored in the uncertainty budget of transferred LNG energy. Thus, the uncertainty in transferred LNG energy can be obtained from the uncertainty in transferred volume, LNG density and SCV.

4.1. Uncertainty in tank volume measurement

The LNG tank volume measurement involves: level gauge, level correction, level to volume correction via gauge table and volume correction terms [4,32]. The dominating error in Membrane type tank volume measurement is due to the gauge table, which has a relative standard uncertainty of 0.20% as reported in [32]. It has been shown that, after combining uncertainty contribution from other sources, the unloaded/loaded volume of a single Membrane type LNG tank has a combined standard uncertainty of u_v = 0.265% (k=1) [32].

We did not analyse the uncertainty of unloaded/loaded volume of a Membrane type tank since no improvements have been proposed to the calculations described in [32]. For the estimation of uncertainty in transferred energy, we will therefore use the standard uncertainty value of u_v=0.265% (k=1) for single tank volume measurement as derived in [32].

4.2. Uncertainty in LNG's superior calorific value (SCV)

The LNG's SCV in mass can be obtained by

$$SCV_{mass} = \frac{\sum_i X_i\, SCV_i}{\sum_i M_i\, X_i} \quad (6)$$

where X_i is the mole fraction of each LNG component, SCV_i is the molar SCV of each component and M_i is the molecular weight of each component.

Reference calorimeters for natural gas have been developed to provide high-accuracy measurements of SCV [8,9,33,34]. The SCV of pure methane can be measured with standard uncertainty of 0.025% (k=1) as reported in [6,7]. The SCV for other components of LNG is assumed to have the same measurement uncertainty as methane [4, 5].

Table 7 shows the composition of a typical LNG sample B (as given in Table 1), with the molecular weight and molar SCV of each LNG component. The SCV in mass for this LNG sample is calculated using (6) to be 54.184 MJ kg^{-1}.

We have used the GUM approach for the propagation from the uncertainty of each LNG component's composition and the uncertainty of each component's molar SCV (SCV_i) to the uncertainty of LNG's SCV in mass through the sensitivity coefficients C_i [22]. The measured composition of each component (for LNG sample B) has a relative standard uncertainty of 0.32% and the molar SCV for each component has a relative standard uncertainty of 0.025% (k=1). The sensitivity coefficient C_i for each source uncertainty component is derived using (6) and shown in Table 8. The uncertainties of molecular weight M_i is negligible, hence they are not considered in the SCV uncertainty estimation. Table 8 shows the standard uncertainty of LNG's SCV in mass is derived to be $\sqrt{\sum_i C_i^2\, u_i^2} = 0.011$ MJ kg^{-1}, thus the relative standard uncertainty of LNG SCV in mass is u_{SCV} = 0.011/54.184 = 0.020% (k=1).

Table 7: The composition of a typical LNG sample B (as given in Table 1), the molecular weight and molar SCV of each component [4, 5] and the calculated LNG's SCV in mass.

LNG Component	mole fraction (%), X_i	molecular weight, M_i (g/mol)	$X_i M_i$ (g/mol)	SCV_i (kJ/mol)	$X_i * SCV_i$ (kJ/mol)	LNG's SCV in mass (MJ/kg)
methane	85.341	16.043	13.6913	891.56	760.87	
ethane	7.898	30.070	2.3749	1562.14	123.38	
propane	4.729	44.097	2.0853	2221.10	105.04	
n-butane	0.992	58.123	0.5766	2879.76	28.57	
iso-butane	0.854	58.123	0.4964	2870.58	24.51	
n-pentane	0.089	72.150	0.0642	3538.60	3.15	
iso-pentane	0.097	72.150	0.0700	3531.68	3.43	
nitrogen	0.000	28.014	0.0000	0	0	
Molecular weight of LNG (g/mol)			19.359			**54.184**

4.3. Combined uncertainty in LNG energy transfer

The total transferred LNG energy is estimated to have an combined relative standard uncertainty given by

$$u_{energy} = \sqrt{u_\rho^2 + u_v^2 + u_{SCV}^2 + 2r_{12}u_\rho u_{SCV}} \qquad (7)$$

where $u_\rho = 0.097\%$, $u_v = 0.265\%$, $u_{SCV} = 0.020\%$ and r_{12} is the correlation coefficient between u_ρ and u_{SCV} [22]. Since the LNG's composition uncertainties affect both u_ρ and u_{SCV}, we assume the correlation coefficient $r_{12} = 1$ (fully correlated between u_ρ *and* u_{SCV}) to get the worst-case uncertainty of the transferred LNG energy. The uncertainty due to tank's unloaded volume can be assumed uncorrelated with the uncertainty of LNG density since the temperature measurement uncertainty of 0.5 K has negligible effect on the estimation of tank's volume.

Table 8: Propagation of the relative uncertainties in composition and SCV_i of each component to the standard uncertainty of the LNG's SCV in mass, for the LNG sample B shown in Table 7.

Source of uncertainty component	**Uncertainty value, u_i**	**Units for u_i**	**Sensitivity coefficient, C_i**	**Units for C_i**	**Contribution to uncertainty for SCV in mass (MJ kg^{-1}), $C_i u_i$**
composition of methane	0.0027	mol mol^{-1}	1.15	MJ kg^{-1} /(mol mol^{-1})	0.0031
composition of ethane	0.00025	mol mol^{-1}	3.47	MJ kg^{-1} /(mol mol^{-1})	0.00088
composition of propane	0.00015	mol mol^{-1}	8.70	MJ kg^{-1} /(mol mol^{-1})	0.0013
composition of n-butane	0.000032	mol mol^{-1}	14.00	MJ kg^{-1} /(mol mol^{-1})	0.00044
composition of isobutane	0.000027	mol mol^{-1}	14.47	MJ kg^{-1} /(mol mol^{-1})	0.00040
composition of n-pentane	0.0000028	mol mol^{-1}	1.90	MJ kg^{-1} /(mol mol^{-1})	0.0000054
composition of isopentane	0.0000031	mol mol^{-1}	1.92	MJ kg^{-1} /(mol mol^{-1})	0.0000060
Molar SCV of methane	0.22	kJ mol^{-1}	0.044	mol g^{-1}	0.0098
Molar SCV of ethane	0.39	kJ mol^{-1}	0.0041	mol g^{-1}	0.0016
Molar SCV of propane	0.56	kJ mol^{-1}	0.0024	mol g^{-1}	0.0014
Molar SCV of n-butane	0.72	kJ mol^{-1}	0.00051	mol g^{-1}	0.00037
Molar SCV of isobutane	0.72	kJ mol^{-1}	0.00044	mol g^{-1}	0.00031
Molar SCV of n-pentane	0.88	kJ mol^{-1}	0.000049	mol g^{-1}	0.000043
Molar SCV of isopentane	0.88	kJ mol^{-1}	0.000050	mol g^{-1}	0.000044
Combined standard uncertainty of SCV in mass (*k*=1), MJ kg^{-1} (SCV in mass: 54.184 MJ kg^{-1})					0.011
Relative standard uncertainty of SCV in mass (*k*=1), u_{SCV}					0.020%

Table 9 summarises the uncertainty budget of LNG energy in custody transfer (for a single Membrane type tank). The expanded relative uncertainty in transferred energy is obtained as U_{energy} = 2 u_{energy} = 0.58% (k=2). We have used the ratio $(u_i)^2/(u_{energy})^2$ to show the percentage of each uncertainty contribution factor (u_i) in Table 9. The dominating contribution is from the tank's unloaded volume measurement uncertainty, which has a percentage of 83.8%.

Table 9: Uncertainty budget for transferred LNG energy

Uncertainty component	Probability distribution	Relative standard uncertainty contribution, u_i (%)	Contribution percentage, $(u_i)^2/(u_{energy})^2$
Tank's unloaded volume, u_v	normal	0.265	83.8%
LNG density estimation, u_ρ	normal	0.097	11.1%
LNG's SCV in mass, u_{SCV}	normal	0.020	0.5%
Correlation effect between u_ρ and u_{SCV}		0.062	4.6%
Combined relative standard uncertainty for transferred LNG energy, u_{energy} (%)		**0.290**	
Expanded relative uncertainty for transferred LNG energy (*k*=2), %		**0.58**	

5. Discussion

The standard uncertainty in LNG density is estimated to be 0.097% after considering various contributing factors and more accurate reference data. This estimated uncertainty value is lower than the value of 0.23% (k=1) given in [4], which shows that the density can be obtained with lower uncertainty.

The uncertainty for LNG composition analysis shown in this paper is lower than that reported in [3], since we have considered the case which uses calibration gas mixture with lower uncertainty and GC with better analysis repeatability. In [3], a typical LNG composition relative uncertainty range is 0.5% - 31% for LNG components, but a good GC can measure the natural gas composition with relative standard uncertainty of 0.3% (k=1) with high-accuracy reference gas mixtures [28].

The estimated standard uncertainty in the LNG's SCV is 0.020%, which is same as the lower bound of the estimated standard uncertainty range for SCV (0.020% to 0.035%) given in [4]. The uncertainty in LNG's SCV caused by composition uncertainty has been studied using a propagation through sensitivity coefficients [22]. This approach is different from that adopted in [3], where the random error for SCV is found to be within ($-B_1$, B), where B is the error bound for positive side, B_1 is the absolute value for the error bound of the negative side. In [3], the maximum error in SCV was estimated to be $e_s = B+B_1$, which was used to estimate the standard uncertainty for SCV, but this approach has over-estimated the standard uncertainty in SCV.

We have used the latest measurement uncertainty of SCV of methane from the GERG reference calorimeter [7], which is 0.05% (k=2) . This uncertainty is about half of the value

(0.11%) used in [3]. The expanded measurement uncertainty of SCV for methane is about 0.11% for the value used in ISO 6976 [4, 5]. The standard uncertainty for the LNG's SCV is estimated to be 0.020%, which is much lower than the standard uncertainty value (0.075%) derived in [3].

The expanded measurement uncertainty in LNG energy transfer is estimated to be 0.58% (k=2) in this paper, which has considered the worst-case scenarios in estimated uncertainties in LNG density and SCV with full correlation between them (both are affected by the composition uncertainties). This result is smaller than the expanded uncertainty's upper bound (1.08%) derived in [3] which has assumed full correlation between unloaded volume, density and SCV. Our estimated expanded uncertainty in transferred LNG energy (0.58%) is smaller than the expanded uncertainty's upper bound (0.74%) reported in [4].

The expanded uncertainty in LNG energy transfer can be further reduced to 0.19% if the unloaded LNG volume is measured more accurately by photogrammetry with an expanded uncertainty of 0.1% [35–37] and LNG temperature is measured with an expanded uncertainty of 0.2 K [4,38], as shown in Table 10 (k=2). The standard uncertainty in LNG density reduces to 0.059% if an expanded uncertainty of 0.2 K is achieved in LNG temperature measurement [4,38].

Table 10: Uncertainty budget for transferred LNG energy if unloaded volume is measured with an expanded uncertainty of 0.1% [35–37] and LNG temperature is measured with an expanded uncertainty of 0.2 K [4,38].

Uncertainty component	**Probability distribution**	**Relative standard uncertainty contribution, u_i (%)**	**Contribution percentage, $(u_i)^2/(u_{energy})^2$**
Tank's unloaded volume, u_v	normal	0.050	28.5%
LNG density estimation, u_ρ	normal	0.059	40.0%
LNG's SCV in mass, u_{SCV}	normal	0.020	4.6%
Correlation between u_ρ and u_{SCV}		0.049	27.0%
Combined relative standard uncertainty for transferred LNG energy, %		**0.094**	
Expanded relative uncertainty for transferred LNG energy (k=2), %		**0.19**	

6. Conclusion

We have studied the uncertainty in LNG density calculation using different calculation models. Four different methods have been used to calculate the LNG density for 5 types of LNG samples with different compositions, with comparison to the measured densities reported in literature. The reported density deviations using EOS-LNG and GERG-2008 methods in literature have also been evaluated to estimate the modelling error limit. We have investigated other factors affecting LNG density estimation, including reference densimeter, sampling/vaporisation effect, composition analysis, temperature measurement and pressure measurement. The uncertainty in LNG's SCV has been analysed with consideration of composition measurement uncertainties and components' SCV uncertainties.

The expanded uncertainty in the transferred LNG energy during LNG loading and unloading custody transfer is estimated to be 0.58% after considering the uncertainties in unloaded volume of a Membrane type tank, LNG density, SCV and their correlation effect. The new approach used in this paper can provide more reliable uncertainty estimation for LNG density, SCV and transferred energy.

The expanded uncertainty in LNG energy transfer can be further reduced to 0.19% if an expanded uncertainty of 0.1% is achieved for unloaded LNG volume measurement by photogrammetry [35–37], along with an expanded uncertainty of 0.2 K for LNG liquid temperature measurement [4,38].

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