Fuel 185 (2016) 932–940

Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

Full Length Article

Novel algorithm for calculating the methane number of liquefied natural gas with defined uncertainty

Björn Gieseking *, Andrew S. Brown

Environment Division, National Physical Laboratory, Teddington, Middlesex TW11 0LW, United Kingdom

HIGHLIGHTS

- Novel algorithm developed for calculating the methane number of liquefied natural gas.
- Algorithm shows good agreement with existing methods for calculating MN.
- The algorithm also determines the uncertainty associated with the calculated MN.
- Relative expanded uncertainty in MN values vary between 0.3 and 0.8%.
- Reporting the uncertainty in MN significantly aids interpretation of the result.

ARTICLE INFO

Article history:
Received 19 May 2016
Received in revised form 14 July 2016
Accepted 27 July 2016
Available online 20 August 2016

Keywords:
Methane number
Algorithm
Liquefied natural gas
Defined uncertainty

ABSTRACT

Liquefied natural gas (LNG) is increasing in importance both as an energy carrier and as a transport fuel. While the developments for an improved infrastructure for LNG are significantly advanced, no commonly agreed method for the characterization of LNG mixtures in terms of the so-called methane number (MN) exist. In this work we present a novel, simple and robust algorithm for calculating the methane number from LNG composition. It combines the detailed experimental data used to develop the commonly used method by AVL (“Anstalt für Verbrennungsmotoren Prof. H. List”) with automated calculation and optimisation routines that guarantee for a high degree of repeatability and reproducibility. This is in accordance with other modern MN calculation tools. The algorithm shows good agreement with other popular methods for a set of exemplar LNG mixtures covering a broad MN range between 60 and 99. Our comparison indicates that the observed differences between the methods might stem from different approaches used for the higher hydrocarbon and inert gas corrections. For the first time to our knowledge the algorithm also determines the uncertainty associated with the calculated MN yielding expanded uncertainties that vary between 0.2 and 0.7 MN depending on the composition of the mixture. We believe that incorporating the uncertainty associated with the calculation of the MN is important for developing a legislation for LNG quality as it would significantly enhance the confidence provided by the results of the calculation tools. In this context the definition of reasonable uncertainty limits in addition to a lower MN limit is recommended.

1. Introduction

Liquefied Natural Gas (LNG) is becoming an increasingly important source for supplying the world with energy. The trade of LNG has increased almost five times since 1990 and is expected to significantly grow further within the next ten years [1,2]. The share of natural gas in the global primary energy mix is 21% [3] with LNG contributing with approximately 10% to the overall gas demand [4]. It accounts for around 30% of international gas exports [5] and the supply has grown faster than any other gas source – at an average 7% per year since 2000 [4]. LNG is considered as more economical alternative to pipeline gas in the case of long distances and is also used to transport natural gas to locations where no pipeline infrastructure exists. Furthermore, assuming an expected increase in the global gas demand of 2.8% per year [3], the contribution of LNG in the energy mix is expected to grow since the existing pipeline infrastructure cannot meet the projected increased demand.

A second important application of LNG arises from its use as a clean fuel for medium- and long-distance transport. In particular for trucks and ships LNG is a real alternative to diesel fuel due to

* Corresponding author.
E-mail address: bjoern.gieseking@npl.co.uk (B. Gieseking).

http://dx.doi.org/10.1016/j.fuel.2016.07.105
0016-2361/

a comparably low pollutant emission while exhibiting a higher energy density than compressed natural gas [6]. LNG has great potential to help achieving the European Commission's targets for greenhouse gas reduction and increased air quality and is therefore part of the EU clean fuel strategy [7]. The growth rate of natural gas as a transport fuel is expected to grow by more than 10% per annum from 2011 to 2040 [8] and it will emerge as main transport fuel within the next years [4,9].

While the development of an infrastructure for LNG as a transport fuel is gaining momentum, the lack of a commonly agreed method to characterise LNGs in terms of the methane number (MN) [10] is impedimental for this progress. The MN is the natural gas counterpart of the octane number and describes the knocking resistance of a gas mixture to engine knocking. It is used by engine manufacturers as a parameter to describe the quality of a natural gas mixture. The MN is defined as the percentage of methane in a methane/hydrogen mixture which has the same knocking behaviour as the gas mixture to be investigated under well-defined testing conditions. Although MN is widely used to validate the quality of LNG mixtures and methods are being written into standards [11], there currently is no single commonly used method for calculating it.

In the present work we describe a simple and robust novel method to calculate the MN of LNG mixtures from their composition. It uses the detailed original data set from the experiments carried out by "Anstalt für Verbrennungsmotoren Prof. H. List" (AVL) [12]. The fully automated calculation and optimisation routines guarantee a high degree of repeatability and reproducibility of the calculated MN. The developed algorithm shows very good agreement with other popular methods based on the AVL data for a set of exemplary LNG mixtures covering a wide MN range: the comparison indicates a linear correlation between the newly developed algorithm and the other methods used for the comparison. Importantly, in addition to the actual MN value, our new algorithm calculates the uncertainty associated with the calculation of the MN based on the use of the ternary diagrams from the AVL study. To our knowledge this is the first time that the uncertainty resulting from the calculation of the MN is explicitly considered for a calculation tool. Including the uncertainty however is crucial when comparing the results of different methods – in particular for determining the quality of a LNG mixture with respect to a defined lower MN limit.

In this paper we first describe the relevant correlations for the development of the algorithm – in particular the AVL method – before explaining the working principle of the new algorithm as well as the calculation of the associated uncertainty in detail. The newly developed method is then validated by comparing it with two other popular methods based on the original AVL data using a set of exemplary LNG mixtures. We then discuss our results and comment on the applicability and limitations of the new algorithm.

2. Theory and methods

2.1. Relevant correlations

The novel algorithm described in this report is based on the detailed experiments carried out and the correlation developed by the AVL in Graz between 1964 and 1970. The studies have been published by the "Forschungsvereinigung Verbrennungskraftmaschinen E. V." between 1968 and 1971 [12]. Important amendments to the AVL correlation have been introduced by "Motorenwerke Mannheim" (MWM), now "Caterpillar Energy Solutions GmbH" [11]. The MWM method is used as an important comparison to validate the newly developed algorithm. Therefore, in order to get a better understanding of the working principle of the developed algorithm, the two aforementioned correlations – the AVL and the MWM method - are briefly described in the following sections. A comparison of the key features of the AVL and MWM methods and the new algorithm can be found in Table 1 while a list of similar notations used to describe the calculation of MN is summarized in Table 2.

2.1.1. AVL method

The AVL method uses a detailed data set summarized in triangular-shaped ternary mixture diagrams (see Fig. 1 as an example). The data was measured in the previously mentioned study using a specific test engine under defined operating conditions, so it is important to note that in order to reproduce the data or add additional measurements the same engine under the exact same operation conditions would have to be used. A different engine or even slightly altered operation conditions of the original engine would yield different results in terms of the absolute MN – therefore called service methane number (SMN). Since the original test engine is no longer available most of the existing methods for calculating the MN are mainly based on the original AVL data.

In order to make use of the ternary diagrams of the AVL study, the composition of the mixture has to be specified in percentage volume fraction and this initial gas mixture has to be divided into sub-mixtures. Before dividing it, however, a reduced mixture is calculated by summing up the volume fractions of *iso*-butane, pentanes (*iso* and *normal*) and higher ($C_{n>5}$) hydrocarbons to the *n*-butane content while ignoring the carbon dioxide and nitrogen content. The resulting mixture is then normalised to 100% volume fraction. This normalised mixture is usually divided into two (maximum three) sub-mixtures with the compositions methane/ethane/butane and methane/propane/butane, respectively, if possible. The partial mixtures are again normalised to 100%

Table 1
Comparison of the main features of the AVL, the MWM and the newly developed methods.

Feature	Methods		
	AVL	MWM	NPL
Reducing mixture	i-C_4H_{10} & $C_{n>4} \rightarrow n$-C_4H_{10}; ignore CO_2 & N_2	As AVL + weighting factors for pentanes and $C_{n>5}$	As AVL
Defining sub-mixtures	Recommendations for sub-mixtures (usually 2)	Selection rules for sub-mixtures	2 sub-mixtures
Determining MN_i	Use of ternary diagrams (manual interpretation)	Calculation using polynomial equation	Calculation based on ternary diagrams
Optimizing ΔMN	Changing composition of sub-mixtures until ΔMN < 5	Minimising ΔMN with numerical procedure	Minimising ΔMN with numerical procedure
Inert gas correction	Only if CO_2 > 2% and/or N_2 > 9%; MN″ from ternary diagram (manual reading)	Always applied; MN″ calculated for CH_4/CO_2 binary	Always applied; Calculation based on ternary diagram assuming CH_4/N_2 binary
Calculation MN	Calculated according to Eqs. (1) and (2)	As AVL	As AVL