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COST-EFFICIENT MEASUREMENT OF ENERGY CONTENT OF PROPANATED BIOMETHANE

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ABSTRACT

This paper presents a correlative method for the cost-efficient measurement of the energy content of propanated biomethane, which is relevant for injection into gas distribution grids. The gross calorific value and Wobbe index were predicted by the regression models from the measured set of physical properties of the gas sample, including speed of sound, sound attenuation parameter, and carbon dioxide concentration. The required properties can be measured using standard sensors and instruments; therefore, they enable in situ application of the method. The results of the experimental validation corroborated with the results of chromatographic analysis.

Keywords: Calorific value, ultrasonic measurement, speed of sound, non-dispersive infrared absorption.

NOMENCLATURE

Abbreviations

CO_2	carbon dioxide
CV	calorific value
GC	gas chromatography
SOS	speed of sound
SAP	sound attenuation parameter
UFMG	ultrasonic flow meter for gases
WI	Wobbe index

Symbols

CV_G	gross CV
WI	WI
c	SOS
α	SAP
x_{CO2}	CO_2 concentration
func	developed regression model

1. INTRODUCTION

Typically, biomethane consists of methane (95-97%) and inert gases such as carbon dioxide (CO_2) and nitrogen (up to 3-5%). Biomethane can be directly utilized in most gas usage applications, including for injection into gas distribution grids. However, in some cases, various local regulations and supply contract limitations restrict the direct injection of biomethane with lower calorific value (CV) or Wobbe index (WI) values than those required. In such situations, the CV (or WI) of biomethane should be increased. Typically, this is accomplished by adding propane to biomethane. This approach is currently used in Sweden for biogas injection into gas distribution grids [1]. The energy characteristics of biomethane at the injection point are typically measured for financial settlement purposes. A common method of gas analysis is gas chromatography (GC), which is accurate but has limited capability of real-time measurement and high operation costs.

As an alternative to GC, correlative methods allow for the measurement of gas energy characteristics at a considerably lower cost [2]. These methods utilize measurements of the physical properties of gas mixtures instead of conducting composition analyses.

In this study, a correlative method to determine the energy characteristics of propanated biomethane based on the measurement of properties of a gas sample using cost-efficient and market available sensors was explored. Two characteristics relevant to the injection of biomethane into gas grids were considered: gross CV and WI. The required measurable properties were the speed of sound (SOS), sound attenuation parameter (SAP), and CO_2 concentration. The proposed approach allows the development of a cost-efficient instrument or even in situ application based on available on-site instruments.

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2. METHODOLOGY DESCRIPTION

The desired gas characteristics were predicted using regression models, from measured physical properties for an unknown gas mixture, as follows:

$$CV_G(or\ WI) = func(c, \alpha, x_{CO2}), (1)$$

where CV_G is gross CV, *WI* is WI, *c* is SOS, α is SAP, x_{CO2} is CO_2 concentration, and *func* is a developed regression model.

The development of the regression model requires the availability of a sufficient quantity of the data for dependent (target) and independent (predictor) variables. In this study, the dependent variables (CV and WI) were calculated from the gas composition under standard conditions (0° C, 100 kPa) for a volumetric basis [3]. The predictor variables were the experimentally obtained data sets of the measurable physical properties of the gas samples: concentration of CO_2 and SOS and SAP under reference temperature and pressure conditions (25° C, 110 kPa).

2.1 Propanated biomethane

The typical compositions of propanated biomethane are shown in Table 1 [2]. Table 1 also presents the maximum and minimum composition ranges observed at the Trelleborg injection point.

Table 1. Typical composition of biomethane and component variations for propanated biomethane as observed at the Trelleborg injection point

Component	Typical	Trelleborg, Sweden	
		Minimum	Maximum
Methane, %	90.94	85.7	98.4
Nitrogen, %	0.69	0.2	0.89
Carbon dioxide, %	2.68	0.6	2.48
Propane, %	5	2.6	8.62
Butane, %	0.5	0.0	0.26
Other, %	0.19	≤0.1	≤0.1

2.2 Measurable properties

The concentration of CO2 could be measured using various technologies, with nondispersive infrared being the most prominent one [2,4].

The SOS in gases depends on the composition and thermodynamic conditions and could be predicted using theoretical models. SOS could also be accurately measured using special sensors or a typical a typical ultrasonic flow meter for gases (UFMG) [4,5].

A sound wave propagating through a gas medium weakens due to the attenuation effects of the medium. This attenuation depends on the composition of the gas mixture. Sound attenuation is of great interest for gas characterization, as it can be potentially measured in combination with SOS [6,7]. However, the measurement of the sound attenuation coefficient usually requires a specific setup [8].

In this study, we considered SAP, which is closely related to sound attenuation in gases and required a relatively simple measurement setup consisting of a single pair of ultrasonic transducers placed opposite each other. Such a setup is typical for UFMGs operating in the time-of-flight technique. A UFMG allows the ultrasound signal dampening of a pass-through impulse to be measured. The proposed approach does not allow for the isolation of the acoustical attenuation coefficient from other effects, including the interaction of the transducer materials with the gas medium and gas flow effects. However, if the technical parameters of the measurement setup are fixed, the only factors affecting the ultrasound signal dampening would be the acoustic impedance and acoustic attenuation of the gas medium [9]. These parameters are defined by the gas composition and thermodynamic state [6]. If the measurement conditions are kept constant, the measured SAP will depend only on the gas composition.

3. EXPERIMENTAL INVESTIGATION

3.1 Experimental setup

The physical properties of propanated biomethane were measured using an experimental rig consisting of a gas mixing unit, sensor array, and GC as the reference analyzer. The gas mixing unit allows the premixing of gas samples into the required gas composition from pure gases (methane, propane, nitrogen, and carbon dioxide). After the mixing, the gas composition was verified using GC, with the measurement uncertainty lower than 0.05% vol.

The mixed gas sample was supplied to the sensor array where the concentrations of CO_2, SOS, and SAP were measured. The measurement was done at a constant flow rate of 0.5 l_n/min (typical for gas analytical instrumentation). The pressure of the gas sample in the measuring system was elevated and kept constant at 110 kPa (abs) in order to avoid any effects due to atmospheric pressure variation (the measurement setup has an open end). The CO_2 concentration was measured using a S-AGM Plus 1031 sensor (Sensor Europe). The measurements of SOS and SAP were accomplished in a heated enclosure with regulated temperature to keep the gas sample at a constant temperature of 25 °C. The temperature of the gas sample was measured with 1/10 DIN probes.

SOS and SAP were measured using a prototype of a typical residential UFMG operating with the single path time-of-flight technique. Two pairs of transducers with different operation frequencies were utilized: 500 kHz and 1 MHz. The transducers were equipped with matching layers optimized for operation in a gas medium. The transducers' excitation and recording of the pass-through signals were performed with the measurement system OPBOX (Optel).

The SOS was derived from the up and down propagation times of the ultrasonic pulses and known separation distance between the transducers [5]. The SAP was obtained as an averaged ultrasonic signal dampening of the pass-through impulse. In particular, the gain level required to maintain the peak of the pass-through amplitude at the desired value was recorded, while the transducer excitation amplitude was kept constant. No diffraction correction was applied to the recorded amplitude due to the negligible effect it had on the utilized transducers and frequencies [8].

The physical properties of a gas matrix representing propanated biomethane were measured. The gas matrix was prepared via an enumeration of all possible variations of gas composition at a 1% vol. step for each subcomponent, as shown in Table 2, with methane as the balance gas. The resulting gas matrix consisted of 131 gas samples. Each gas sample was measured twice, including interim purging with nitrogen, and two detached data sets were obtained: training and testing.

Table 2. Component range of the gas matrix

Parameters	Minimum value	Maximum value
Methane	84%	100%
Nitrogen	0%	2%
Carbon Dioxide	0%	3%
Propane	0%	10%
CV	37.7 MJ/m^3	45.8 MJ/m^3
WI	49.1 MJ/m^3	56.8 MJ/m^3

3.2 *Measured SOS and SAP*

The results of the SOS measurement showed a favorable response to the concentrations of the subcomponents and dependency of the response on ultrasonic frequencies. A discrepancy with the theoretical values obtained from the GERG-2008 model was also observed. This was well expected for a polyatomic gas mixture such as propanated biomethane due to the molecular relaxation effects for the utilized ultrasonic frequencies [7].

Figure 1 shows the recorded and normalized SAP values for the test gas matrix. Normalization was

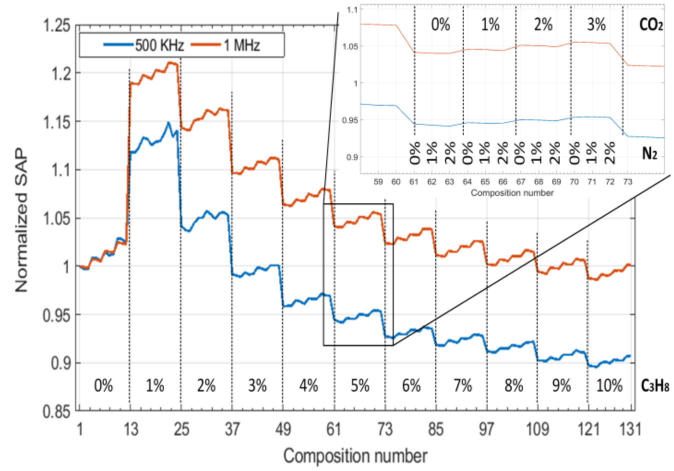

Fig 1 Normalized SAP for the studied gas matrix

performed according to the readings of pure methane for each frequency to simplify comparison. The SAP parameter shows favorable sensitivity to the propane content in the biomethane gas mixture within the studied ranges. The response has an evident frequency dependency due to the molecular relaxation effects of ultrasonic frequencies. This in general agreement with the results reported by other studies [7,8]. The SAP response to CO_2 concentration is considerably lower but still evident; no frequency dependence was observed. SAP showed no usable response to the nitrogen content in biomethane for the studied composition ranges and utilized instrumentation.

3.3 *Development of regression model*

The training data set was used for the development of the regression model according to (1). Several regression techniques have been tested using the MATLAB Regression Learner module, with standard settings. The best performance was achieved via the Gaussian process regression (Table 3).

Table 3. Performance indicators of the developed regression models with the data set used for training

Absolute Error	CV, MJ/m^3		WI, MJ/m^3	
	Max	Avg	Max	Avg
500 kHz	0.928	0.255	1.181	0.402
1 MHz	0.246	0.048	0.346	0.088

4. RESULTS AND DISCUSSION

Experimental validation was performed on the test data set. Figure 2 shows the CV prediction performance for 500 kHz and 1 MHz transducers.

The results of the CV and WI prediction for 1 MHz transducers are encouraging. The maximum relative deviation is less than 0.65%. The relative mean absolute deviation is about 0.2%.

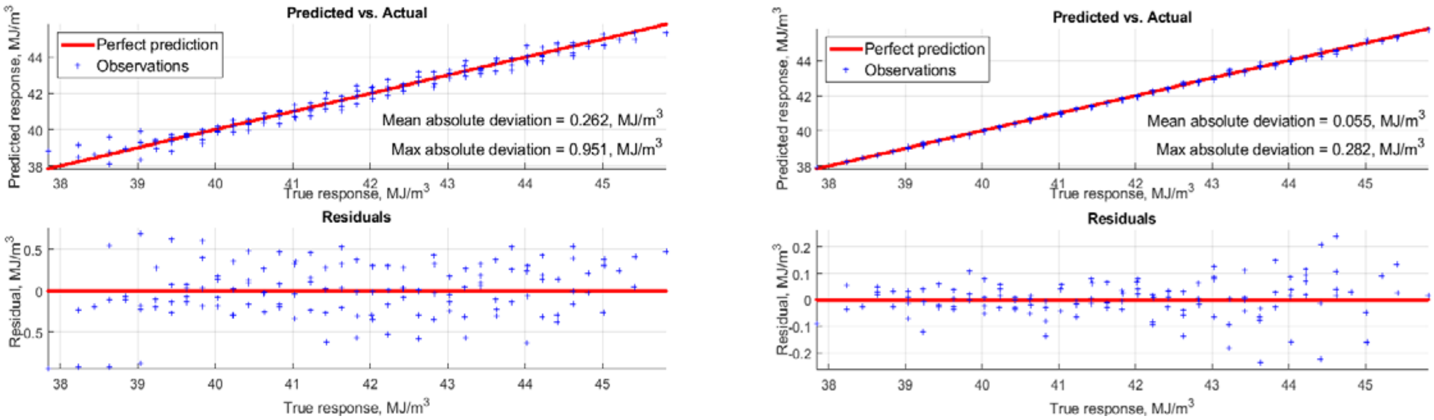

Fig 2 Performance of the developed regression models in the prediction of CV: 500 kHz (left) and 1 MHz (right)

5. CONCLUSIONS

The study shows that SAP in propanated biomethane can be a useful measurable parameter (in combination with other physical properties) for a correlative method for the determination of CV and WI. The interest in SAP lies in the fact that it could be measured in combination with SOS using one physical sensor. It was shown that the SAP (and consequently, the sound attenuation coefficient) measured at a 1MHz frequency could provide valuable information about a gas sample and be utilized in gas analytical instrumentation together with SOS. This is in particular the case for polyatomic gas mixtures with light hydrocarbons that show strong molecular relaxation effects in range of ultrasound frequencies of 1MHz.

The main advantages of the approach proposed in this study are real-time measurement and low cost of technical implementation. Utilization of standard sensors would allow the in situ implementation of the method. However, the observed dependence of SAP on certain ultrasonic frequencies will lead to different results for other ultrasonic frequencies and measurement setups. The methodology of development of the regression model should be specific for different measurement conditions, transducers, and measurement setups.

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