Taras KOTURBASH [1], **Agnieszka BICZ** [2], Wiesław BICZ [2]
[1] DEPARTMENT OF ENERGY TECHNOLOGY, KTH ROYAL INSTITUTE OF TECHNOLOGY, 68 Brinellvägen, SE-10044 Stockholm, Sweden
[2] PRZEDSIĘBIORSTWO BADAWCZO PRODUKCYJNE OPTEL SP. Z O.O., 30 Morelowskiego, 52-429 Wrocław, Poland

New instrument for measuring the velocity of sound in gases and quantitative characterization of binary gas mixtures

Abstract

A new instrument for precise measurements of velocity of sound in gases was developed and tested. The instrument implements improved velocity of sound measuring technique based on time-of-flight measurement of ultrasonic pulses. The instrument was developed primarily for conducting measurements in natural gas, it can operate in wide ranges of natural gas compositions as well as other gas mixtures with velocity of sound parameter in range of 200 to 500 m/s. The instrument allows conducting measurement of velocity of sound propagation in various gas mixtures with average repeatability of equal to ±1 cm/s at normal conditions. The accuracy of measurement depends on calibration conditions and approach, and has good agreement with theoretically calculated values. The instrument has a modular design of control unit and flow-through measuring chamber. The set of distinctive features and adjustable measuring parameters of the instrument allows further improving of applicability for other gases and gas mixtures and various measurement conditions. The instrument showed good performance in the task of quantitative characterization of binary gas mixtures by velocity of sound parameter.

Keywords: ultrasonic measurement, velocity of sound, speed of sound, natural gas, time of flight, characterization of gas mixture, analysis of gas mixture.

1. Introduction

The velocity of sound (VOS) is the distance travelled per unit of time by a sound wave as it propagates through an elastic medium. VOS varies for different substances, primarily due to difference in elastic properties: sound travels most slowly in gases, faster in liquids and even faster in solids. Therefore, VOS might be considered as univocal physical property of various substances, and if measured, could help to characterize or identify unknown substance.

In non-destructive testing the knowledge of VOS is usually needed for evaluation of thickness or depth of flaw location. The measurement can be based on measurement of time-of-flight (TOF) of ultrasonic pulse and known properties of material or fluid. This approach is also used for characterization of various gases and gas mixtures. VOS in an ideal gas depends only on its temperature and chemical composition. Therefore, VOS could give distinctive information about the gas mixture and is used in various methods for analyses of gas mixtures: measurement gas purity, characterization of binary gas mixtures and even evaluation of properties of multicomponent mixtures like natural gas [1, 2].

In our previous work we have employed measured VOS as input parameter for characterization of combustible and qualitative properties of natural gas [3]. While VOS allowed us to receive specific information about some properties and composition of gas mixture, the question of accurate and inexpensive measurement of it in gases in non-laboratory conditions still remained open. The findings indicated that aimed requirement for accuracy measurement of VOS should be ± (3-5) cm/s, which is rather complicated task for field instrumentation.

In this article we described the improved technique and new instrument for accurate and low-cost measurement of VOS in gases. The developed instrument was tested in wide range of natural gas like mixtures and also used for characterization of binary gas mixtures by VOS parameter under in-flow conditions.

2. VOS measurement in gases

Several approached and techniques for measurement of VOS in gases are known: TOF (pitch-catch and pulse-echo), resonance method, phase change and others. Each approach has its benefits and limitations, especially in relation to costs, accuracy and applicability to various gases and gas mixture ranges.

TOF measurement technique is one of the simplest approaches that are based on direct measurement of time that is needed by the beam of sound to travel the distance between two points. TOF technique is the main basic principle for majority of ultrasonic gas flow meters and has several variations of implementation in the latter. VOS value is usually a sub-product of the measurement conducted by ultrasonic flowmeter and is used for verification of the condition of the meter [4]. The average uncertainty of VOS measurement by ultrasonic flow meter is not lover then ±60 cm/s for in field applications [5]. However, the actual repeatability of VOS measurement results might reach up to 0.03% while difference with theoretical value is relatively big 0.3% due to pressure and temperature uncertainty [6]. In absolute values for pure methane at normal conditions this will mean repeatability at ±13 cm/s, and even bigger discrepancy with theoretical values. The discrepancy with theoretical values is caused mainly by temperature measurement accuracy and rather different intended purpose of the flow meter and therefore other considerations for technical implementation of TOF techniques.

The main benefit of TOF technique is that it is based on electronic components for time measurements. Current state of development of such electronics allows digitizing signals with very high accuracy and resolution. The electronical components themselves are usually offer one-chip-solution with very high performance specifications. In theory, TOF technique can reach also quite extremely high resolution of VOS measurement when higher frequency of the ultrasound is used. But this leads to main drawback of TOF technique when applied in gases: the practically reachable resolution of VOS measurement is limited by strong attenuation of ultrasound that increases proportionally to the square of the beam frequency. Due to this fact the majority of commercially available ultrasonic transducers for air or gas medium have relatively low frequencies (20-500 kHz). Those air- or gas-coupled transducers should also have specific matching layers for minimization of loses at solid/gas medium boundary and should be excited with very high voltage pulses.

The resolution of VOS measurement by TOF technique mainly depends on several factors: selected frequency of transducers, resolution and accuracy of time measurement and travel distance of ultrasonic beam. The properties of gas medium will also affect the level of excitation of ultrasonic pulse and its attenuation along the travel path. Therefore, additional requirements should be set for amplification of received signal and signal to noise ratio at high amplification levels. The combination of above stated factors results in complexity and eventual costs of the implementation of the TOF technique in the final instrument.

In previous attempts we have used TOF pulse-echo measurement technique by combined ultrasonic transducer (resonance frequency at 1 MHz) and concave reflector [3]. Due to influencing factors of temperature fluctuations and high attenuation of ultrasound, the measurement was conducted in slightly pressurized chamber at fixed static conditions of temperature and pressure. This approach allowed us to reach the accuracy of VOS measurement equal ±0.8 m/s. During subsequent experiments we have also discovered major problems with the proposed setup: transducer ringing after exciting and low resolution of time measurement was the reason to use relatively long travel distance between transducer and reflector. This resulted in high attenuation of the ultrasonic beam along the travel path and relatively high dispersion of measurement results within one measurement cycle. The strong impact of temperature and gas fluctuations along the travel path was also inevitable and therefore unit shoved unreliable performance in in-flow conditions.

As the solution for the problem high attenuation of ultrasonic beam in gas we have considered decreasing the travel distance in the first place. This also allowed us to lower the requirements of power levels of exciding pulse and increase signal to noise ratio of received signals. As a trade-off we needed to increase the resolution of time measurement from µs (previously used) to tens of ns to keep up with targeted VOS measurement resolution.

For the development of the new instrument we have also considered the following additional requirements:

- increase sampling rate and add on-the-fly measured data processing for better averaging and prelaminar evaluation of the results of each measurement cycle;
- extend applicability for in-flow conditions for continuous operation in bypass or inline connection of gas line;
- target low manufacturing costs and utilize market available electronic components with standard specifications.

3. Improved VOS measurement technique

Initial experiments have shown that previously selected arrangement of combined transducer and reflector puts considerable restrictions and requirements on transducer design and related electronics. The major problems were electrical and over-voltage protection circuits, long travel distance due to ringing after transducer excitation, specific requirements to the design of combined transducer. In order to minimize the costs associated with solutions of these problems, we have decided to select the arrangement with two transducers, one sending and one receiving.

The arrangement with two opposite placed transducers still allows implementing both TOF techniques: pitch-catch (by pass-through pulse) and pulse-echo (by 1st reflection) (Fig. 1). Due to high attenuation of ultrasonic pulses in gases, the considerable amplification of sending and/or received signals is required. The generation of high power ultrasonic pulse was limited by targeted application of the instrument and related restrictions (safety considerations due to explosive medium of natural gas, low power consumption for continuous operation and low manufacturing costs). It was decided to use higher amplification of received signal in favor of lower level of generating pulse. At inducing pulse of 220 V and travel distance between transducers of 16 mm in methane, the required post attenuation for keeping amplitudes of pass-through signal and 1st reflection at desired level of 300 mV was around 63 dB and 73 dB correspondingly (Fig. 1). For other gases, to receive desired level of signals, even higher amplification levels for each pulse might be required. Therefore, the variable amplification possibilities for pass-through and 1st reflection signals should be also foreseen.

One of conditions of proper TOF measurement is the analysis of various delays in matching layers that might reach several microseconds in total for gas-coupled transducers. After initial testing of the prototype of new instrument and analyses of recorded signals, we have discovered systematic error of TOF1 measurement caused by delay in matching layer and amplification circuit (Fig1). It was also observed that this error could vary for different measurement conditions and amplification levels. This delay is one of the main reasons for uncertainties for VOS measurements in ultrasonic flow meters. This uncertainty could be decreased during precise calibration on pure gases, but still depends on measurement conditions and specific gas mixture properties [7].

To overcome such potential limitation, it was decided to carry out the TOF2 measurement between transmitted pulse and 1st reflection (see Fig. 1). This allowed us to ignore the delay in matching layer and amplification circuits of receiving transducer due to the fact that it is already included in both time measurements: for pass-through and reflected pulses. Target TOF2 of ultrasound propagation between transducers will be obtained by subtraction of time measurement 1 from time measurement 2. VOS value will be calculated from TOF2 divided by traveled distance (the doubled distance between transducers).

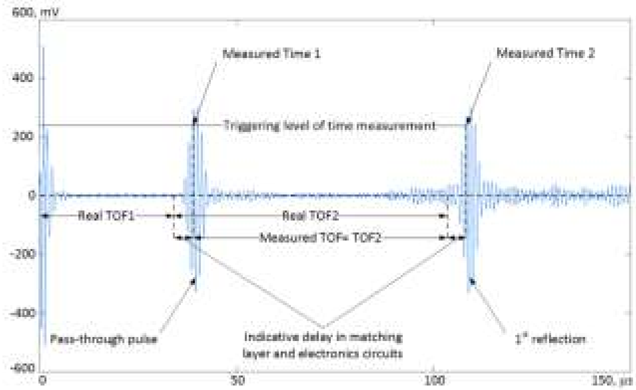

Fig. 1. TOF measurement by pass-through and 1st reflected pulses in pure methane

The most crucial aspect of any TOF measurement is selection of proper triggering criteria for time measurement. For most precise TOF measurement the triggering of time measurement should be identical for both pulses. Most obvious choice of peak amplitudes or 1st front of pulse does not work well for various gases. During initial tests we have observed that peak amplitude of 1st reflection do not always fall on 3rd positive half-wave peak of the signal, as it is usually the case for pass-through signal. In addition to that, the front of 1st positive half-wave of the 1st reflection was usually covered by the noise at high levels of amplification (Fig. 1). However, it was observed in numerous instances (various gas mixtures, levels of exciting pulse and amplification) that the front of 2nd positive half-wave of received pulse is usually distinguishable for both signals and therefore can serve as proper triggering criteria for time measurement.

4. Description of the new instrument

For implementation of improved VOS measuring technique in gases, the new instrument for TOF/VOS measurements was developed by OPTEL. The instrument consists of 2 major functional blocks: the measuring chamber with installed sensors and the control unit opTim (Fig. 2).

The control unit opTim has connections to sending and receiving traducers, two temperature sensors, communication port and 3 pin connection (signal pass-through and external triggering) to OPBOX© for signal visualization. OpTim is a slave mode unit that implements the control command from PC.

OpTim 1v2 has following main features:

- Pulser: 0-400 V, 4 us
- Receiver:
  - input impedance: 1 kOhm
  - preamplifier: 20 dB
  - variable gain amplifier: 0-80 dB
  - bandwidth: 600-1200 kHz (-3 dB)
- The temperature sensors:
  - 2 channels
  - PT1000/PT500, 2-wire
  - meas. method: discharge time measurement
  - precision: up to 0.004°C
- Communication:
  - interface: RS232
  - baud rate: 57600
- Power requirements: 5 V, 0.2 A (1 A max)
- Dimensions: 35×85×183 mm, weight: 330 g.

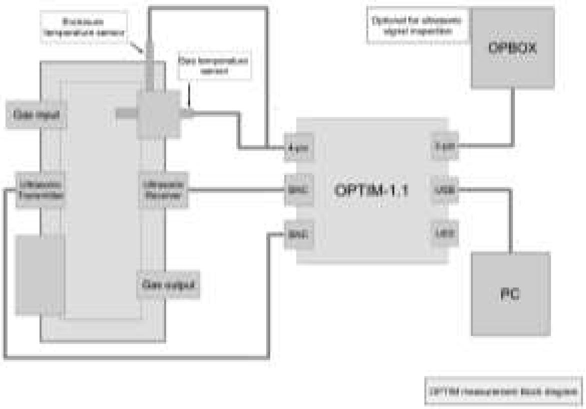

Fig. 2. Functional block diagram of opTim instrument developed by OPTEL

Triggering of time measurements (stop event) was implemented in standard level measurement (LM) mode. Threshold level for stop event could be set by VREF parameter. To get stable TOF readings VREF should be chosen to be above noise level and first period of transducer response. TOF measurements accuracy and stability in this mode strongly depends on amplitude of peaks, their stability and noise. Additionally, any change of VREF threshold level cause minor changes in TOF measurement (Fig. 3).

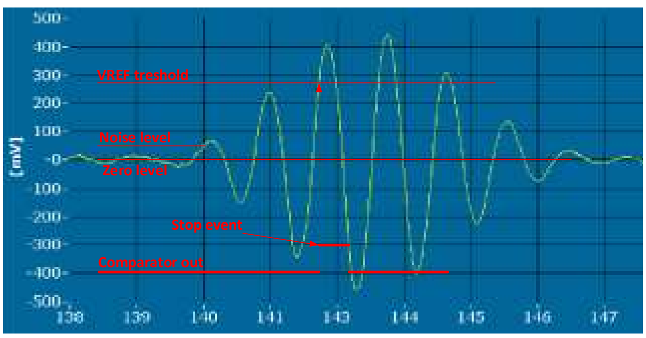

Fig. 3. Time measurement triggering principle of standard LM mode

To get TOF measurements less sensitive to noise, total gain, transducer response shape in certain gas mixtures with higher attenuation levels of ultrasound, special comparator mode was also implemented. It allows generate triggering event at "zero level" with usage of dynamically calculated time mask (Fig. 3). Working at "zero level" of signal makes triggering event much more immune for noise and not sensitive for signal slope shape, distortion and other factors. Sinusoidal waveform near zero have also biggest slew rate – better for sharp and stable comparator response. The information about proper time mask settings is calculated from the transmitted signal and known parameters of measuring chamber. This method delivers stable and accurate TOF measurement results even in gas mixtures with very weak signals and low signal to noise ratio due to attenuation effects.

The measuring chamber has a flow-through design with gas inlet and outlet connections, ports for two ultrasonic transducers placed opposite to each other, two temperature sensors for gas medium and enclosure, and additional port for other sensors (Fig. 3). The chamber consists of two half-parts that are pressure tightly joined by eight screws and separated by isolation made from elastic layer that limits the direct transmission of sound waves through housing. This transmission is significant source of noise in the system and must be reduced. Transducers used in the device were specially developed for the chamber. They employ PZT vibrating in thickness mode at resonance frequency of 1 MHz and specially designed matching layer.

OpTim allows conducting continuous measurement of TOF with preselected sampling and averaging conditions in range of 1..1000 measurements per cycle. Post-processing also includes handling of potential timeouts of TOF measurement, median filter and calculation of standard deviation of TOF values for assessment of measurement performance. The instrument allows on-fly adjustment of amplification and time masks parameters similar to approach employed in typical non-destructive ultrasonic flaw detectors.

The instrument is capable of measuring VOS in various gases in the range of 200-500 m/s. However, this was limited by firmware only due to targeted application in natural gas and could be extended to considerably wider range of 50..1500 m/s with same hardware components.

Fig. 4. Measurement setup: control unit opTim with measuring chamber and OPBOX

5. Experimental testing

Before any measurements of unknown gas mixture by the instrument, the precise distance between transducers should be obtained during calibration on pure gases. The calibration procedure consists of comparing measured VOS (calculated from measured TOF and rough distance between transducers) with reference values for air, methane and nitrogen (or any other calibrating mixtures) at known pressure and temperature. As result, the real distance between transducers is estimated more precisely and furtherly used for calculation of correct VOS value by the instrument. I should be noted that such calibration is valid only for specified pressure range. Further pressure increase inside the chamber might affect the distance between transducer and require additional calibration.

The instrument was tested on pure gases and binary mixtures of methane with other gases found in natural gas: nitrogen, carbon dioxide, hydrogen. Several gas flow conditions were also tested from zero flow up to 1 l_n/min. Tests were done at ambient temperature and pressure. The temperature of the chamber and gas mixture inside it was also recorded for further analysis. The gas mixture flow was loaded on the atmosphere and therefore no pressure deviations inside the chamber were observed during experiments.

The utilized setup and measurement procedure for any gas mixture was the following:

1) Setup the unit with default (or pre-selected) settings and start continuous mode of TOF measurement.
2) Run OPBOX© for signal visualization (Fig. 5).
3) Adjust amplification of STOP1 (pass-through) signal to get the amplitude in range of 0.3-0.33 V
4) Adjust amplification delay for STOP2 signal (1st reflection) to be in middle of pass-through and 1st reflection signals.
5) Adjust amplification of STOP2 signal, similar to STOP1.
6) Set time masks for STOP1 and STOP2 signals between minus 10 us mark of each 1st wave front or as close as possible to expected TOF value and check measured values (Fig. 5).
7) Fine tune all parameters to get stable TOF and lowest stable standard deviation of TOF readings at selected averaging settings.

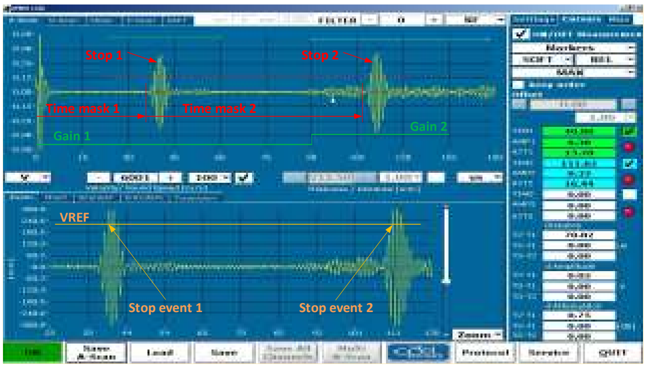

Fig. 5. Signal visualization in OPBOX for binary gas mixture with clarifications to the setup procedure of the instrument. Composition of gas mixture: methane 95.0%, nitrogen 5%

VOS measurement results in pure methane are shown in Fig. 6. VOS value is recalculated by the instrument on the fly based on actual TOF measurement and calibrated distance between the transducers. The results of VOS measurements showed good agreement with theoretical values obtained from GasCalc software according to AGA8 calculation for specified temperature and pressure [8, 9]. The corresponding measured TOF values and standard deviation of measured results are shown in Fig. 7.

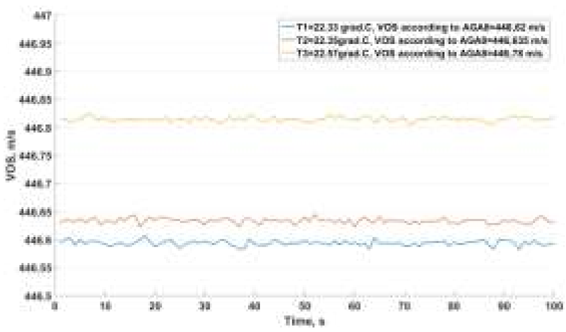

Fig. 6. Measured by the instrument VOS in methane at 3 temperatures, gas flow 0.5 l_n/min, purity N2.5, gas pressure inside the chamber P_{abs}=102.1 kPa

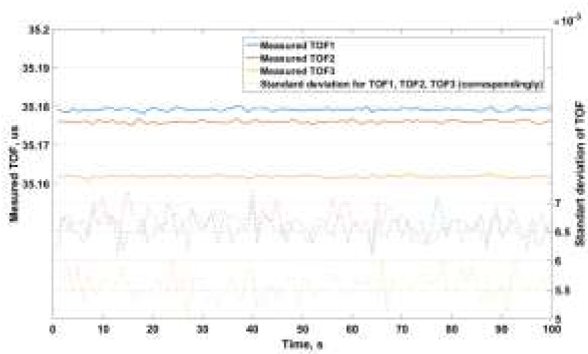

Fig. 7. Measured TOF values and related standard deviations corresponding to VOS measurements in Fig. 6

6. Characterization of binary gas mixtures

It is known fact that VOS in gas mixture depends on the composition of its constituents and is univocal for any macro point of measurement due to uniform distribution of molecules of the components in gas mixture [10]. Therefore, by knowing the relation of VOS from its constituents it is possible to characterize the binary gas mixture by measurement of this single physical property [1, 5].

The instrument was used for quantitative characterization of compositions of binary mixtures of methane with admixture of nitrogen and carbon dioxide in range of 0 to 10% (Fig. 8). The gas mixture was prepared by mixing of pure gases by Bronkhorst Select series mass-flow controllers. The total error in final gas mixture did was lower than 0.6% for main component (methane) and 0.06% for secondary components. The additional reference analysis of received mixture was accomplished by laboratory grade gas chromatograph.

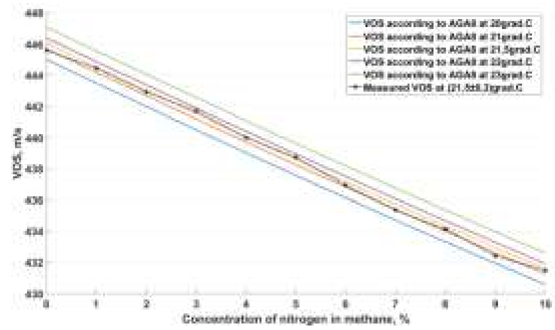

Fig. 8. Measurements VOS in binary gas mixture (nitrogen diluted in methane) plotted against theoretically calculated value of VOS

The VOS value measured by the instrument showed good agreement with theoretical values obtained from AGA8 calculation. Therefore, the instrument could be used for quantitative characterization of binary gas mixture by measured VOS and temperature. The target concentration of the binary gas mixture is obtained from comparison of measured and calculated theoretical values for known pressure and temperature conditions.

Characterization of gas mixtures with more than two constituents is also possible, but it requires additional measurement of those extra components with help of physical properties that are not affected by other constitutes in the gas mixture. For example, NDIR sensor could be used measurement of carbon dioxide concentration in trinary mixture of methane, carbon dioxide and nitrogen because its reading are not affected by last two. For quaternary mixture of methane, carbon dioxide, nitrogen and hydrogen additional two NDIR sensors, one for carbon dioxide and other for methane, could be used.

7. Conclusions

As a part for solving problem of precise VOS measurement in natural gas we have developed and tested the new instrument. The instrument can conduct VOS measurement with high accuracy and resolution of ±1 cm/s in no-flow and up-to 1 l_n/min flow conditions for technological process monitoring.

We have showed that the new instrument could be used for characterization of composition of unknown gas mixture (by VOS parameter). Arbitrary binary gas mixtures could be characterized by the instrument' measurement of VOS value. In the next step of development we will try to add measurement of attenuation of ultrasound beam that will deliver additional information about the gas mixture with only one physical sensor.

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Taras KOTURBASH, PhD (Cand. Tech. Sc.)

A graduate of IFNTUOG in Ivano-Frankivsk, Ukraine, Department of Technical Diagnostics and Monitoring (2013). Specialty: methods and instruments of control and determination of internal structure of materials, non-destructive testing and evaluation. Currently: PhD Student at Department of Energy Technology at KTH Royal Institute of Technology in Stockholm, Sweden.

e-mail: koturbas@kth.se

Agnieszka BICZ, Ms, eng.

Graduate of Technical University in Wroclaw, department Electronics and Telecommunication, since 1993 works at R&D laboratory of OPTEL Sp. z o.o. where she is developing advanced industrial and laboratory systems for quality control (for non-destructive testing) and support of technological processes, based on ultrasonic technology. Has made significant experience with NDT systems and biometrics.

e-mail: A.Bicz@optel.pl

Wiesław BICZ, MSc, eng.

Graduate of Technical University in Wroclaw with a specialty coherent optics. Inventor and founder of PBP Optel. Since 1985 he is working in the area of ultrasonic technology, with special application in finger recognition.

e-mail: W.Bicz@optel.pl