This paper concerns speed of sound measurements performed in three different gas mixtures at constant temperature and pressure while the concentration of the gases was varied. The performed experiments used an ultrasonic, sing-around, gas flow meter equipped with silicon based transducers. The center frequency of the transducers was 800 kHz. Speed of sound was measured in mono-, di- and triatomic gases: argon (Ar), oxygen (O₂) and carbon dioxide (CO₂), in either air or nitrogen (N₂) as a background gas. The gas under investigation was mixed with the background gas in a test chamber and the concentration of the gas under examination was varied between 0% and 100%. A gas chromatograph was used in order to accurately determine the composition of the gas mixture. The experiments show that measured speed of sound, as a function of gas composition, agrees with the speed of sound obtained from theory. The achieved data also show that the speed of sound measurements was performed with low standard deviation. Thus, one can conclude that this type of ultrasonic gas flow meter is well suited in determining gas concentration in a binary gas mixture as well as flow velocity. The technique could be of value in both industrial and medical applications.

**Introduction**

A regular ultrasonic transit-time flow meter has the inherent capability of performing simultaneous measurements of both flow velocity and speed of sound in the medium flowing through the meter. The speed of sound in a gas depends on the composition of the gas as well as on its temperature and pressure. By using the speed of sound information from the flow meter, it is potentially possible to determine the composition of the gas flowing through the meter. The problem of determining the gas composition in a gas mixture by acoustic methods has been a field of research for decades. Various techniques has been invented and investigated, like e.g. phase shift methods, acoustic interferometry methods and pulse techniques. In the pulse technique, the propagation time is measured for a pulse sent by one transducer to a second transducer at a fixed distance. From that, the speed of sound in can be determined. This technique has successfully been used in binary gas composition measurements by Hallewell et.al. [1], Joos [2] and others. The problem of determining gas composition by acoustic methods has recently been addressed by Dain & Lueptow [3], [4] who used ultrasonic spectroscopy in determining the composition of three-gas mixtures. The method used, utilizes the spectral content of the ultrasonic pulse paired with molecular vibration relaxation time modeling of the gas mixture, a rewarding but quite complex technique. In this paper, we explore the possibilities of using a regular, transit-time, ultrasonic flow meter under atmospheric pressure and room temperature in determining the composition of a binary gas mixture.

**Theory**

Classically, the speed of sound in an ideal gas could written as $c_0 = \left(\frac{\gamma R_0 T}{M}\right)^{1/2}$. Now, if we consider a mixture of ideal gases, the expression above could be rewritten. By assuming that the specific heat ratio $\gamma$ is written as the mole weighted specific heat ratio for the mixture and that the molecular weight of the mix $M$ is the mole fraction weighted sum of the mole weights of the constituents as
\[ y = \frac{\sum_{i}^{n} x_i C_{pi}}{\sum_{i}^{n} x_i C_{vi}} \] (1)

and

\[ M = \sum_{i}^{n} x_i M_i \] (2)

In (1) above, \( C_{vi} \) is the specific heat at constant volume and \( C_{vi} = C_{pi} - R_0 \). Using (1) and (2) in the equation for the speed of sound we arrive at

\[ c_0^2 = R_0 T \frac{\sum_{i=1}^{n} x_i C_{pi}}{\sum_{i=1}^{n} x_i M_i (C_{pi} - R_0)} \] (3)

where \( R_0 \) is the universal gas constant, \( T \) the absolute temperature, \( x_i \) the mole fraction of the \( i \):th component, \( M_i \) the molecular weight and \( C_{pi} \) the heat capacity at constant pressure. In the experimental setup used in this study, temperature and gas concentration, i.e. mole fraction, is simultaneously measured. The speed of sound in the gas mixture is then determined by equation (3).

**Experimental method and materials**

The meter used in the experiments presented in this study is an ultrasonic gas flow meter equipped with silicon-based ultrasound transducers, which have a center frequency of 800 kHz. The flow meter uses the well-known sing-around technique and detailed descriptions of the technique could be found in e.g. Lynnworth [5]. For the meter, one can derive an equation for the speed of sound in the air flowing through the meter based on the up- and down-stream propagation times for the ultrasonic pulses, i.e.

\[ c = \frac{L}{\frac{t_1}{2} + \frac{1}{t_2}} \] (4)

where \( c \) is the speed of sound, \( L \) is the distance between the transducers and \( t_1 \) and \( t_2 \) are the up and down-stream propagation times.

A general error propagation analysis, Coleman and Steele [6], is performed on (4). We then obtain the uncertainty limits on \( c \) from the following expression.

\[ \delta_c = \left[ \frac{3}{2} \sum_{i=1}^{3} \frac{\partial c}{\partial \xi_i} \delta \xi_i \right]^{1/2} \] (5)

where \( \delta_c \) is the uncertainty limit on speed of sound. \( \xi_i \) represents the \( i \):th variable and \( \delta \xi_i \) is its uncertainty limit. The same type of uncertainty analysis is also performed for the speed of sound, equation (3). The uncertainties for the different parameters are shown in table 2. The uncertainty limit range for the speed of sound obtained from the flow meter is found to be \(| \delta_c | = 0.54 \sim 0.57 \) m/s, or \(~ 0.16 \) %, at 95% confidence level. The most significant source of error for \( \delta_c \) is found to be the uncertainty in the distance between the transducers. The standard deviation on speed of
sound values obtained from the flow meter is found to be of at least one order of magnitude smaller than the uncertainty stemming from the distance between the transducers.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Value</th>
<th>Uncertainty $\delta x_j$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$L$</td>
<td>61.4 mm</td>
<td>± 0.1 mm</td>
</tr>
<tr>
<td>$t_1, t_2$</td>
<td>~ 0.178·10^{-3} ms</td>
<td>± 3.3·10^{-8} sec</td>
</tr>
<tr>
<td>$T$</td>
<td>~ 20 °C</td>
<td>± 0.5 °C</td>
</tr>
</tbody>
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TABLE 1. Uncertainty values for the ultrasonic gas flow meter and equation (3).

From the uncertainty analysis of equation (3) we find that the uncertainty limit $|\delta c_0| = 0.59 - 0.14$ m/s or $\sim 0.16 - 0.04 \%$. The sources of uncertainty for the uncertainty limit $\delta c_0$ are temperature and mole fraction values but the only source of uncertainty considered in this study is temperature. The uncertainty limit $\delta c_0$ could be reduced considerably by using a temperature sensor with better precision than the thermometer used in this study.

Three gases were investigated at room temperature and atmospheric pressure. Argon (Ar) and oxygen (O$_2$) were mixed with nitrogen (N$_2$) as a background gas and their mole fractions ranged from 0% to 100%. Carbon dioxide (CO$_2$) was mixed with air as a background gas and the carbon dioxide mole fraction ranged from 0.072% to 3.148%. All the experiments were performed in the temperature range between 19 °C to 20 °C. Values on the specific heat for each gas at constant pressure was calculated for different temperatures using empirical relations from Moran & Shapiro [7].

<table>
<thead>
<tr>
<th>Background gas</th>
<th>Gas mixed with background gas</th>
<th>Range of mixing, Mole fraction [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air</td>
<td>CO$_2$</td>
<td>0.072 – 3.148</td>
</tr>
<tr>
<td>N$_2$</td>
<td>O$_2$</td>
<td>0 – 100</td>
</tr>
<tr>
<td>N$_2$</td>
<td>Ar</td>
<td>0 – 100</td>
</tr>
</tbody>
</table>

TABLE 2. Background gases and the gases they are mixed with. The background gas is mixed with an increasing amount of the mixing gas during the course of the experiment. All gases used in the experiments were of laboratory grade.

For the measurements of speed of sound in the gases mentioned above, a custom made chamber of Plexiglas was used. The chamber was equipped with an inlet for the gas under investigation, an outlet where samples of the gas mixture were drawn and two water locked outlets. This enables the pressure in the chamber to be kept constant and very close to the ambient atmospheric pressure. The sample outlet was connected to a gas chromatograph that analysed the composition of the gas mixture in the chamber. The flow meter was mounted on the inside of the chamber lid, which sealed hermetically to the chamber body. A small fan was placed in the chamber in order to create flow through the flow meter as well as mix the gas under investigation with the background gas. The temperature of the gas mixture was monitored using a thermometer that was placed inside the chamber. The chamber was initially filled with the background gas. The gas under investigation was during the experiment allowed to enter the chamber at a very low flow rate and to be mixed with the background gas.

A gas chromatograph, a Varian – Chrompack Micro GC CP-2003P, was used in performing the analysis of gas samples that were taken from the gas chamber. Before each experiment, the gas chromatograph was calibrated using calibration gases. During the experiments, the gas chromatograph and the gas flow meter recorded concurrent measurements every third minute on gas composition, flow velocity and speed of sound.
Discussion of results
The figures 1 to 3 show the speed of sound as a function of gas concentration, or mole fraction. Both experimental and theoretical values on the speed of sound in the gas mixtures are plotted. It is seen that the experimentally obtained values agree well with the values obtained from calculations using equation (3). One can also observe that the experimentally obtained speed of sound for the investigated gases argon, oxygen and carbon dioxide at 100% concentration corresponds well to literature data on speed of sound at room temperature and atmospheric pressure, see Kaye & Laby [8], Lide [9].

FIGURE 1. Speed of sound in nitrogen (N₂) – argon (Ar) mix as a function of argon concentration. The line represents speed of sound calculated from equation (3) and the circles represent experimental data. The temperature is 19.5 ± 0.5 °C.

FIGURE 2. Speed of sound in nitrogen (N₂) – oxygen (O₂) mix as a function of oxygen concentration. The line represents speed of sound calculated from equation (3) and the circles represent experimental data. The temperature is 19.3 ± 0.5 °C.
FIGURE 3. Speed of sound in air – carbon dioxide (CO₂) mix as a function of carbon dioxide concentration. The line represents speed of sound calculated from equation (3) and the circles represent experimental data. The temperature is 19.2 ± 0.5 °C.

Conclusion
Speed of sound was measured in mono-, di- and triatomic gases; argon (Ar), oxygen (O₂) and carbon dioxide (CO₂) mixed with in either air or nitrogen. It was found that the measured speed of sound show good agreement with data obtained from theory. It is concluded that by simultaneous measurements of temperature and speed of sound, the ultrasonic flow meter could be applied in determining the mole fraction of the constituents in flowing binary gas mixtures. The largest sources of uncertainty are found to be the temperature measurement of the gas and the measurement of the distance between the two transducers in the flow meter. Employing a more precise temperature measurement and a better calibration procedure for the meter are essential to reduce these uncertainties. The investigated measurement technique could be of value in applications where flow and mixing ratio of gases are important, for instance, in monitoring of gas combustion processes or breathing-air monitoring for anaesthetics or other medical purposes.

References