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REAL-TIME COMPOSITION DETERMINATION OF GAS MIXTURES

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ABSTRACT

In this paper we describe the development of a model and system to analyse the compositions of gas mixtures up to 4 components. We present measurements with binary and ternary mixtures, on compositions with components that range over an order of magnitude in the value for the measured physical properties. The system is consists of a coriolis, density, pressure and thermal flow sensor. With this system it is possible to measure the viscosity, density, heat capacity and flow rate of the medium. We demonstrate that if the properties of the individual components are known, binary mixtures can be analyzed within 1 % and ternary within 2 % accuracy.

KEYWORDS

Multi Parameter, gas mixtures, thermal flow sensor, coriolis flow sensor.

INTRODUCTION

In flow dosing systems there is a demand to not just measure or control flowrate, but also measure the type or composition of the flow. For instance in biogas production; here the measurement of methane and inert gas fractions can give information about the energy content and in this way serve as a quality control [1-5]. A second example is purging of reaction chambers in semiconductor industry. Many processes here involve toxic or explosive gasses. Before equipment, that contains these gasses, can be opened, it should to be properly purged. A real-time monitoring system of gas composition can help to indicate threshold levels for harmful gasses and signal when it is save to open the reaction chamber. In this way reduce costly down time of equipment and aid in operator safety. Here we show that by combining a medium independent Coriolis mass flow measurement with thermal and pressure sensors, we can extract physical properties of the medium. In a second calculation step, the composition can be calculated based on these measured medium properties

Model

The model presented in figure 1 links a pressure, density, thermal and Coriolis sensor [5]. The latter one acts as a medium independent mass flow sensor. The thermal sensor gives an output that is proportional to both the mass flow and heat capacity of the medium. Linking the two sensors yields an independent measurement of the heat capacity. Secondly, with an independent measurement of the medium density, the volume flow can be determined by combining density and mass flow. This finally gives the viscosity when the pressure needed to drive the volume flow through a fixed flow obstruction is measured in the laminar flow regime.

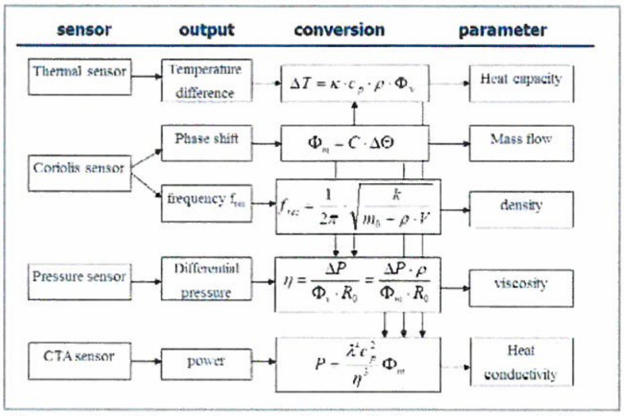

Figure 1: Schematic overview of the sensors, their relations and their transfer function.

When a gas mixture is composed of a limited number of components, the measured physical properties can be used to analyse this composition.

Formula one illustrates how the total heat capacity is a sum of the contribution of the two components. Since the total fraction is one, this equation can be solved and yield the compositing if the heat capacity of the two components. In the formula c_p, c_{p1}, c_{p2}, φ_1 and φ_2 are respectively the total heat capacity, the heat capacity of component 1 and two and the mass fractions of the two components.

$$cp = cp_1 * \varphi_{m1} + cp_2 * \varphi_{m2} \qquad (1)$$

Formula 1 can be expanded to include all the parameters measured in our system. When written as a function of the volume fraction and put into matrix form, this yields equation 2

In formula 1 and 2 cp, φ_m, φ, η and ρ are respectively the heat capacity at constant pressure, mass fraction, volume fraction, viscosity and the density. The subscripts indicate mixture components.

$$\begin{pmatrix} \rho_1 & \rho_2 & \rho_3 & \rho_4 \\ \eta_1 & \eta_2 & \eta_3 & \eta_4 \\ 1 & 1 & 1 & 1 \\ \frac{c_{p1}}{\rho_{tot}} & \frac{c_{p2}}{\rho_{tot}} & \frac{c_{p3}}{\rho_{tot}} & \frac{c_{p4}}{\rho_{tot}} \end{pmatrix} * \begin{pmatrix} \varphi_1 \\ \varphi_2 \\ \varphi_3 \\ \varphi_4 \end{pmatrix} = \begin{pmatrix} \rho \\ \eta \\ 1 \\ c_p \end{pmatrix} \qquad (2)$$

It becomes clear from formula 2, that with our system we are able to analyse gas mixtures of up to four components. For this analysis it is necessary that the physical constants of the components are known, and that at least one constant makes the gas distinguishable from the other components.

Results

A schematic drawing of the setup is shown in figure 2. The coriolis, thermal and pressure sensor are respectively a M13, F-111AC and P-502C all manufactured by Bronkhorst High-Tech B.V. The density meter is a northdome sensor purchased from Avenisense. Mixtures where supplied by controlling the gas flow of the individual components with thermal mass flow sensors at a constant total volume flow of 1 lit/ min. The pressure is measured over the thermal flow sensor, since over this section the flow is in the luminaire Hagen-Poiseuille regime, where there is a linear relation between pressure and volume flow.

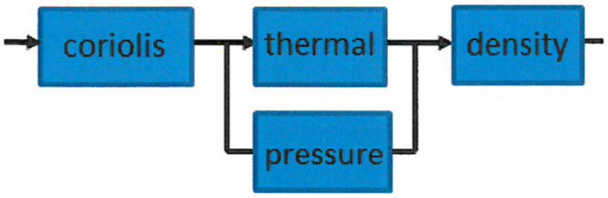

Figure 2: schematic of the measurement setup.

The first result shown in figure 2 gives the volume fraction of an air hydrogen mixture based on the measured heat capacity. The bottom graph in figure 2 shows a result for a methane nitrogen mixture. In the figure both the measured heat capacity as the heat capacity calculated based on the applied flow rates of the components are plotted. The inset of the picture shows the calculated and applied volume flow rate. The calculated fractions are calculated with the aid of formula 1 and converted to volume fractions.

In both cases the volume flow fractions, could be calculated with an accuracy of less than 1 %.

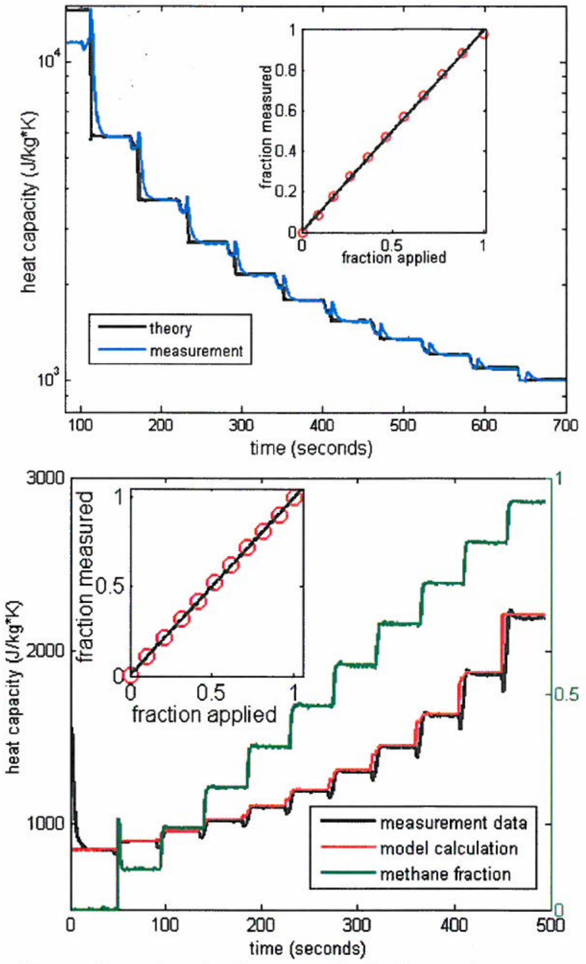

Figure 3: top) mole fraction of a hydrogen/air mixture based on the measured heat capacity. Bottom) mole fraction of methane/nitrogen mixture.

The measured heat capacity shows deviation for 100 % hydrogen, this most likely is due to slight contamination with air, due to the large difference in density and heat capacity the difference between theory and measurement can already be accounted for by a 0.1 v/v % air fraction in the hydrogen flow.

In figure for a measurement result is given for a mixture of hydrogen, air and argon. Since the mixture consists of only three fractions, three equations suffice to yield a solution. Since we measure the mass flow and pressure we can calculate the ratio between the density and viscosity, as can be seen in figure 1. Based on this parameter together with the measured heat capacity we calculate the volume fraction of the three components.

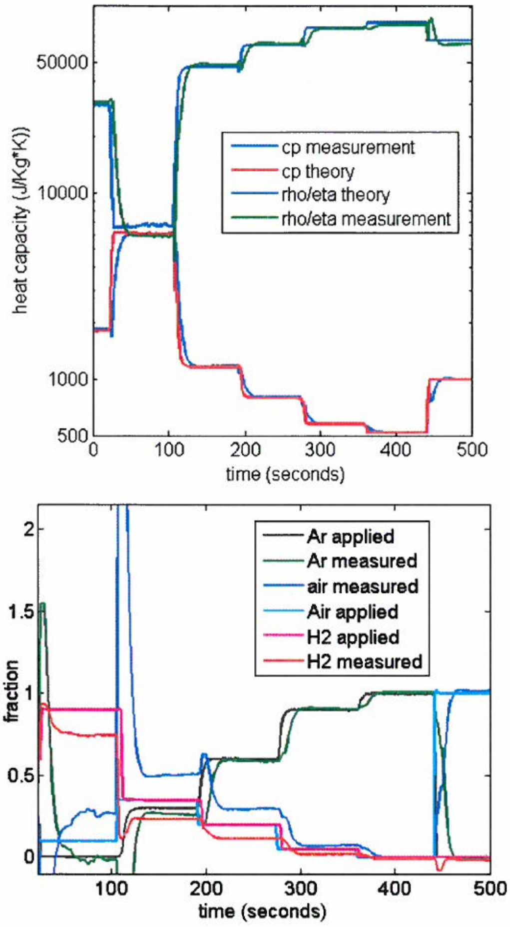

Figure 4: Top) heat capacity and ratio between density and viscosity. Bottom) calculated volume fraction of the three components.

It can be seen in the top figure that for the ratio between density and viscosity as well as the heat capacity, there is a good agreement between the theory and the measurement results. This however, does not result in an accurate calculation of the volume fractions. The result in the bottom graph of figure 4 indicates that there is good correlation between applied and measured fractions, however, with large absolute variations.

The resulting variations are caused largely by selectivity of the measured parameters. It can be seen in the top part of figure 4 the 2 measured parameters are each other's inverse and hence cannot be used adequately to calculate the composition. By incorporating a separate density measurement the measurement can be more specific.

Table 1: gas properties of nitrogen and carbon oxide.

Property/ component	Nitrogen	carbon monoxide	Hydrogen
density	1.150	1.150	0.083
Heat capacity	1040	1043	14300
viscosity	$1.747 \cdot 10^{-5}$	$1.665 \cdot 10^{-5}$	$8.8 \cdot 10^{-6}$
Thermal conductivity	0.02487	0.026	0.176

Table 1 illustrates a limitation of this method. It can be seen that although heat capacity and density are very different for hydrogen and nitrogen, the product is the same. Furthermore it can be seen, that for carbon monoxide and nitrogen all the parameters listed in the table are almost identical. This is caused by the very similar molecular structure and atomic weight. To test if a density measurement can improve the results we have performed tests on a mixture of methane, nitrogen and carbon dioxide. We have selected a mixture of these three components since this resembles the composition of certain types of biogas.

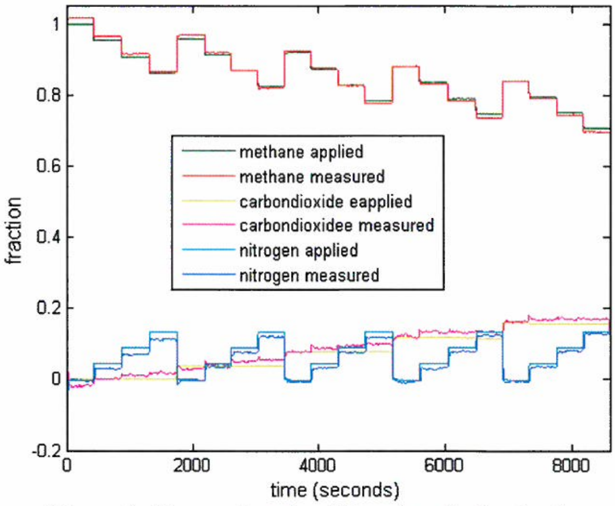

Figure 5: The caption should be placed after the figure.

Conclusions

We have designed and realized a system and model with which we can real-time determine the composition of gas mixtures, if the properties of the components of the mixture are sufficiently distinguishable. The model is based upon a multi-parameter flow measurement system, consisting of a Coriolis and thermal flow sensor, a density meter and a pressure sensor. The system enables direct measurement of flow rate and physical properties of gas mixtures. Binary mixtures have been measured with accuracy of better than 1 %. Ternary mixture has been measured within 2 % for a mixture of methane, nitrogen and carbon dioxide.

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