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APPLICATIONS AND PERSPECTIVES OF ULTRASONIC MULTI-GAS ANALYSIS WITH SIMULTANEOUS FLOWMETRY

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TWO PAGE EXTENDED ABSTRACT

We have developed ultrasonic (“sonar”) instrumentation [1,2] for simultaneous flow and composition measurement in a variety of gas mixtures. Flow and composition are derived from measurements of ultrasound transit times in opposite directions in a flowing process gas.

Continuous, real-time precision measurements of relative concentration of binary pairs of gases are required in many applications. The presence of other gases can however cause ambiguities in the measurement: a particular measured sound velocity can be the result of varying combinatorial concentrations of additional gases.

We have developed a sound velocity based algorithm to compensate - or “deduct” - the effects of additional gases, allowing the concentrations of a pair of gases of primary interest to be acoustically measured on top of a varying baseline from “third party” gases whose concentrations in the multi-gas mixture are measured by other means.

Several instruments are used in the ATLAS experiment at the CERN Large Hadron Collider (LHC) to continuously monitor C₃F₈ (R218) and CO₂ coolant leaks into N₂-purged environmental envelopes surrounding parts of the ATLAS detector [2]. Precision in molar concentration of better than 2.10⁻⁵ is routinely seen in mixtures of C₃F₈ in N₂ in the presence of varying known concentrations of CO₂. Further instruments monitor air ingress and C₃F₈ vapour flow (at high mass flows around 1.1 kgs⁻¹) in a 60 kW thermosiphon C₃F₈ evaporative cooling recirculator.

This instrumentation and analysis technique, targeting binary pairs of gases of interest in multi-gas mixtures, is promising for mixtures of anaesthetic gases, particularly in the emerging area of xenon anaesthesia.

In our instruments two 40 MHz transit time clocks are started synchronously with the leading edge of the first sound pulse transmitted in each direction and are stopped the first above-threshold signal received at the facing transducer. Bi-directional transmission allows simultaneous measurement of the gas flow (via up/down transit time differences) and sound velocity (from their average). Since the sound velocity in a gas mixture at known temperature and pressure depends on the molar concentrations of its components it can be compared in real-time to a stored *concentration vs. sound velocity* database previously generated using data from theoretical models [3] or measurements made in calibration mixtures.

The gas analysis algorithm is based on the generalized equation for sound velocity, c , in a gas:

$$c = \sqrt{\frac{\gamma RT}{M}} \quad (1)$$

where R is the molar gas constant (8.314 J·mol⁻¹·K⁻¹) and T is the absolute temperature (Kelvin). The adiabatic index γ_m for the mixture is given by the ratio of the weighted sums of

molar specific heat at constant pressure (C_{p_i}) to that at constant volume (C_{v_i}) of the n components:

$$\gamma_m = \frac{C_{pm}}{C_{vm}} = \frac{\sum_i w_i C_{p_i}}{\sum_i w_i C_{v_i}} \quad (2)$$

where w_i are the molar fractions of the components ($i = 1 \rightarrow n$). Similarly, the molar mass, M , of the mix (kg·mol⁻¹) is given by the weighed sum of the component molar masses, M_i :

$$M = \sum_i w_i M_i \quad (3)$$

so that equation (1) may be recast as:

$$c = \sqrt{\frac{\sum_i w_i C_{p_i} RT}{\sum_i w_i C_{v_i} RT}} \quad (4)$$

Figure 1 illustrates as an example the variation of sound velocity with the concentration of C₃F₈ (molecular weight = 188) in N₂ (m.w. = 28) in the range 0 → 1%; a gas pair and concentration range of particular interest in the ATLAS cooling application.

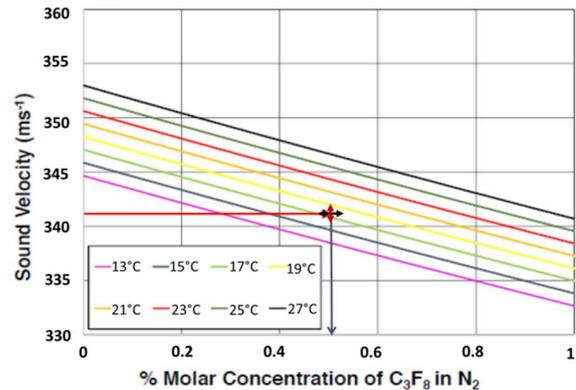


Figure 1. Variation of sound velocity with concentration of binary C₃F₈/N₂ mixtures in the range 0→1% C₃F₈ at 1 bar_{abs}.

The added horizontal and vertical indicators illustrate the relationship between the uncertainties in precision of the measured sound velocity, δc , and the corresponding mixture determination, $\delta(mix)$. At any concentration of the two components;

$$\delta(mix) = \frac{\delta c}{m} \quad (5a)$$

where m is the local slope of the *sound velocity vs. concentration* curve ([ms⁻¹]/%).

For clarity in Fig. 1 the ordinate and abscissa are reversed. In operation the % concentration is inferred from the measured sound velocity. At any measured sound velocity the corresponding uncertainty on the mixture is then given by

$$\delta(mix) = \delta c * m' \quad (5b)$$

where m' is local the slope of the *concentration vs. sound velocity vs. curve* (%/[ms⁻¹]).

In the example of Fig. 1 the average slope of the *sound velocity vs. molar concentration* curve is around -12.25 ms⁻¹ per % of C₃F₈ in the range of interest of 0→1% molar C₃F₈.

The $\pm 0.025 \text{ ms}^{-1}$ uncertainty in sound velocity $\hat{\alpha}$ in our instruments [2] results in a corresponding mixture uncertainty of $\pm 2 \cdot 10^{-5}$. Over this narrow range the variation of sound velocity with C_3F_8 concentration can be fitted as linear, requiring only two fit parameters.

In other applications with a wider concentration range of the two principal gases, higher order polynomials are required. For example in xenon-oxygen based anaesthesia (0 - 80% Xe in O_2) requires a 5th order polynomial (fig. 2). The average gradient over the range is around 0.52% per $\text{m}\cdot\text{s}^{-1}$. From eq. (5b) we see that a target precision of $\pm 0.1\%$ Xe in O_2 implies a sound velocity measurement precision of $\pm 0.19 \text{ m}\cdot\text{s}^{-1}$.

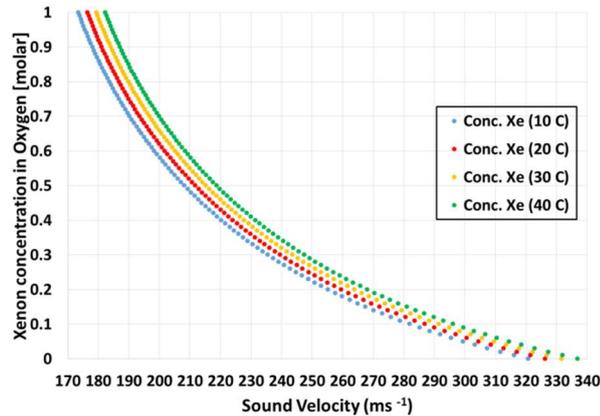


Figure 2. Composition/sound velocity dependence in xenon-oxygen mixtures at several temperatures (1 bar_{abs}) [4].

The gas mixture analysis database contains sets of polynomial fit parameters from curves of composition vs. sound velocity curves for a gas pair of primary interest calculated from eq. (4) using thermodynamic data (C_p , C_v) [3] on a grid covering the expected process temperature, pressure and the expected concentration ranges of *all* known gases present in the mixture.

Figure 3 illustrates the algorithm for 3-stage interpolation of parameters of sound velocity vs. $\{\text{C}_3\text{F}_8 \text{ in } \text{N}_2\}$ composition generated and stored in intersection “cans” (•) on a 3-D grid of $\{T, P, \text{ppm } \text{CO}_2\}$ covering 13-25°C (0.5°C step: $n_T = 25$ points); 900-1100 mbar_{abs} (20 mbar step: $n_P = 11$ points), and 0-10000 ppm (1000 ppm step: $n_{\text{CO}_2} = 11$ points). The 8 nearest grid points in $(T, P, \text{ppm } \text{CO}_2)$ space are chosen to define the smallest cuboidal volume encompassing the process measurables $\{P_{\text{meas}}, T_{\text{meas}}, \text{CO}_{2\text{meas}}\}$. For clarity these 8 points are represented by their 12 projections on the (P, T) , $(P, \text{ppm } \text{CO}_2)$ & $(\text{ppm } \text{CO}_2, T)$ facets visible in Fig. 3 (a).

In practice, since the temperature, pressure and CO_2 concentration (all measured simultaneously with the acoustic transit times) will fall between the grid values: a multi-step interpolation is used. With three process parameters, the interpolation is cubic (fig 3). Clearly, if no third-party gas is present, the interpolation is reduced to a 2-step process starting with 4 grid points in $\{T, P\}$ space. For each additional process parameter the database acquires an additional dimension, and can become tesseract or penta-ract (4 or 5 process parameters respectively), or higher-dimensional. Figure 4 illustrates measurements of C_3F_8 concentration seen in 4 N_2 -purged environmental zones sequentially monitored in a 16-hour supercycle by aspiration through a single instrument. These volumes have varying permeability to CO_2 ingress from the exterior. CO_2 concentration is monitored

with an infrared monitor¹ allowing unambiguous C_3F_8 measurement on top of a varying known CO_2 concentration. During the measurement period shown in fig. 4 the CO_2 concentration in one zone underwent a step increase due to an external manipulations while the C_3F_8 leak rates into each zone (due to small coolant leaks) remained fairly constant.

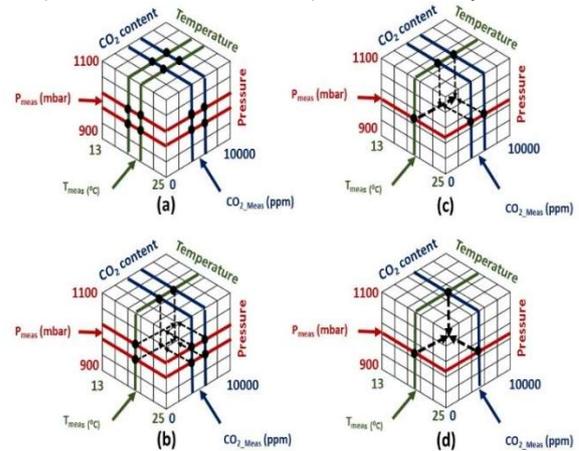


Figure 3. The 3-step interpolation of sound velocity vs. $\{\text{C}_3\text{F}_8 \text{ in } \text{N}_2\}$ concentration fit parameters held in “cans” (•) at the nearest stored grid intersections to determine those corresponding to the measured $\{T, P, \text{CO}_2 \text{ contamination}\}$ process conditions.

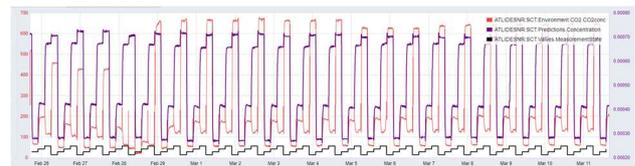


Figure 4. Variations of % molar C_3F_8 (violet) and ppm CO_2 concentrations (red) seen in 4 sequentially-monitored N_2 -purged zones surrounding the ATLAS “SCT” sub-detector (zones 1-4 indicated by “valve state”: black).

Although ultrasonic gas analysis is primarily seen as a binary gas analysis tool, we have demonstrated that it can be successfully used with higher order gas mixtures if the concentration of the additional component(s) is known from other sources and the reference database uses their thermodynamic data in the approach of eqns (2-4) and fig. 3. This analysis approach has potential in various industrial and medical applications where simultaneous flowmetry and accurate real-time analysis of a gas pair of primary interest are required in a multi-process gas environment. One such example is in the emerging field of xenon-based anaesthesia, where ultrasonic instrumentation can simultaneously monitor flow and xenon concentration to a precision better than 0.1%, and can help improve the post-operative recovery of expensive xenon gas for recycling.

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¹ GE Telaire infra-red sensor: 10000 ppm full scale output.