

Sonic Gas Analysis

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In the late-1800s, Germans used a “methane whistle,” known as a *Schlagwetterpfeife*, to determine the presence of hydrogen or methane in the air that was extracted from underground mines. A modern version of that sonic gas analysis concept is described that should be suitable for safety-related applications in garages that shelter hydrogen or methane-powered vehicles or monitoring cook stove pollutants. A compact, self-resonant standing wave tube is capped at both ends by two small electrodynamic speakers. The cylindrical copper resonator has two slits at the mid-plane to allow a fan to continuously sample the air which is brought to the resonator’s temperature after being drawn along the resonator body. Variation in the resonator’s temperature is compensated electronically by an integrated-circuit temperature sensor.

Historical Introduction

In the late-1800s, one of the world’s most famous (and controversial) German chemists, Fritz Haber, invented the “methane whistle,” known as a *Schlagwetterpfeife*, to determine the presence of hydrogen or methane in the air that was extracted from underground mines [1]. Since both methane ($M_{CH_4} = 16.04$ kg/kmole) and hydrogen ($M_{H_2} = 2.01594$ kg/kmole) have molecular masses that are substantially less than the molecular mass of dry air ($M_{Air} = 28.9644$ kg/kmole), their presence in otherwise pure air cause the sound speed in the resultant gas mixture to increase so that $c_{mix} > c_{air}$. Haber passed the gas mixture through a whistle that acts as a flow-excited, open-open resonator whose fundamental resonance frequency f_1 is given by the ratio of the sound speed in the mixture c_{mix} to the wavelength λ of the sound,

$$f_1 = \frac{c_{mix}}{\lambda} = \frac{c_{mix}}{2L_{eff}} \cong \frac{c_{mix}}{2L}. \quad (1)$$

The inequality in Eq. (1) is motivated by a small end-correction that makes the effective length of the whistle $L_{eff} > L$, where L is the length of the whistle. Since $c_{mix} > c_{air}$, the pitch of the whistle increased if either methane or hydrogen were present. By 1884, these methane whistles also began to appear in the United Kingdom [2].

The first acoustical gas analyzers that employed electronic detection were developed in the 1920’s [3, 4]. A version based on tube electronics was patented in 1942 [5]. These analyzers proved to have a very high sensitivity, accuracy, and a rapid response time. Additionally, the acoustical analyzers had the advantage of being able to work under a variety of conditions (temperature, pressure, flow rates, etc.).

Although the “methane whistle” was used in mine-safety applications early in the 20th century [6], the rise of precision electronic instrumentation, in particular, the Wheatstone Bridge and galvanometer, made sensors that produced changing electrical parameters (*e.g.*, electrical resistance) in response to changes in the desired measurand more common in all fields of instrumentation. For determination of gas mixture concentration, measurement of the mixture’s thermal conductivity using the resistance difference between two hot-wire thermal conductivity gauges (one in the unknown mixture and the other in a “reference gas”) became the most common non-specific sensor system for determination of gas mixture concentration [7].

The rise of gas analyzers based on the gas mixture’s thermal conductivity halted most development of the sonic gas analysis method for nearly a half-century, until the early 1970’s, when a bit of a renaissance was initiated by the use of sound speed measurements to determine isotopic ratios of ^3He to ^4He in superfluid helium research [8, 9]. Other examples of the return of interest in the sonic technique are a detector of air leakage into a helium recovery system [10], as well as four other instruments for measuring the isotopic ratio of ^3He - ^4He mixtures [11, 12, 13, 14].

In the late 1990’s, Zipser [15] developed an acoustic gas sensor to measure the humidity in exhaust air from industrial driers. Like some of its predecessors, that sonic gas analyzer included two thermally coupled resonators: one filled with a reference gas and the other with the gas mixture to be analyzed. The acoustic gas sensor proved to be reliable under extreme conditions. Zipser, *et al.* [16, 17], also described an acoustic method for ternary gas analysis *via* simultaneous measurements of isentropic and isothermal sound speed in the mixture.

In 1998, Golden, Garrett, and Keolian, presented the design of a dual-differential resonator for acoustical gas analysis at a joint meeting of the Acoustical Society of America and the Congress on Acoustics in 1998 [18]. That same resonator strategy was used for the experiments described in a thesis by Horta for bacterial metabolic rate measurements [19]. Subsequent to the publication of this open-open dual-differential resonator topology, a patent on this concept (which did not include the location of the microphones to suppress mode-hopping) was (erroneously) issued to Karr [20].

Using an analyzer with a single resonator, Schuhmann used acoustical gas analysis to make non-invasive determination of gaseous diffusion rates through porous media [21, 22]. This technique, with its higher accuracy and precision, provided new insights into the determination of tortuosity factors and generated corrections to the standard chromatographic analysis which produced results that were previously confounded by advection and non-equimolar effects [23].

The flow-through sonic gas analyzer that will be described here differs from previous applications of the sonic gas analysis technique in its implementation of the transduction mechanisms, in its thermal equilibration of the gas and measurement of gas temperature, and in the compatibility of the resonator with a compact scheme for forced-convective sampling of the atmosphere.

Sound Speed in Gas Mixtures

The speed of sound in a gas mixture c_{mix} depends upon the mean molecular mass of the gas mixture M_{mix} , absolute (Kelvin) temperature T , the ratio of specific heat at constant pressure c_p to the specific heat at constant volume c_v , $c_p/c_v = \gamma_{mix}$ (polytropic coefficient), and the Universal Gas Constant [24], $\mathcal{R} = k_B N_A$, which is the product of Boltzmann's constant k_B times Avogadro's Number N_A .

$$c_{mix} = \sqrt{\frac{\gamma_{mix} \mathcal{R} T}{M_{mix}}} \quad (2)$$

If the contaminant gas, or gas mixture, with concentration x , has a mean molecular mass M_2 , and the uncontaminated gas or gas mixture has a concentration $(1-x)$ and a mean molecular mass M_1 , then the mean molecular mass of the mixture is simply their concentration-weighted average:

$$M_{mix}(x) = (1-x)M_1 + xM_2; \quad (0 \leq x \leq 1). \quad (3)$$

Computation of the mean polytropic coefficient for the contaminated gas γ_{mix} is not as simple because, unlike the mass which is an extensive quantity, the polytropic coefficient is an intensive quantity:

$$\gamma_{mix}(x) = \frac{c_p(x)}{c_v(x)} = \frac{c_{p1}(1-x) + c_{p2}x}{c_{v1}(1-x) + c_{v2}x}; \quad (0 \leq x \leq 1). \quad (4)$$

For ideal gases, $c_p - c_v = \mathcal{R}$.

Substitution of Eqs. (3) and (4) into Eq. (2) yields an explicit expression for the sound speed $c(x)$ in terms of the concentration x :

$$c(x) = \sqrt{\frac{[c_{p1}(1-x) + c_{p2}x] \mathcal{R} T}{[c_{v1}(1-x) + c_{v2}x][M_1(1-x) + M_2x]}}. \quad (5)$$

Therefore, if the specific heats of the gases and their molecular masses are known, then the concentration x of gas 2 can be determined from a measurement of the sound speed.

Sensitivity to Contaminant Gas

Resonator Frequency Sensitivity

The resonance frequencies f_n of a resonator are directly proportional to the sound speed of the gas contained in the resonator, as illustrated by the resonance frequency f_1 of the first mode of a whistle provided in Eq. (1). Since the sonic gas analyzer described herein uses the measurement of resonance frequency, it is convenient to express the concentration x of the contaminant (*i.e.*, gas 2) in terms of the resonant frequency $f(0)$ of the resonator which contains the pure, uncontaminated gas 1, and the frequency $f(x)$ measured when the concentration of gas 2 is x :

$$\frac{f(x)}{f(0)} = \frac{c(x)}{c(0)} = \sqrt{\frac{\gamma(x) M_1 T(x)}{M(x) \gamma_1 T(0)}}. \quad (6)$$

In many applications, it is most important to determine small amounts of the contaminant gas 2 present in a previously uncontaminated gas 1. For that case, we define a sensitivity $\beta = \delta f/f(0)$, where $\delta f = f(x) - f(0)$, and $x \ll 1$. Logarithmic differentiation of Eq. (6) yields,

$$\frac{\delta c}{c(0)} = \frac{\delta f}{f(0)} = \frac{1}{2} \left[\frac{\delta \gamma}{\gamma_1} - \frac{\delta M}{M_1} + \frac{\delta T}{T(0)} \right]. \quad (7)$$

Expressing $\delta M = (M_2 - M_1)x$, the resonator's relative frequency shift $\delta f/f(0)$ can be expressed in terms only of the gas specific heats and molecular masses, and the temperature change δT :

$$\frac{\delta f}{f(0)} = \frac{1}{2} \left[\frac{c_{p2} - c_{p1}}{c_{p1}} - \frac{c_{v2} - c_{v1}}{c_{v1}} - \frac{M_2 - M_1}{M_1} \right] x + \frac{\delta T}{2T(0)}. \quad (8)$$

The temperature-independent portion of the sensitivity β , can therefore be written as

$$\beta = \frac{1}{2} \left[\frac{c_{p2} - c_{p1}}{c_{p1}} - \frac{c_{v2} - c_{v1}}{c_{v1}} - \frac{M_2 - M_1}{M_1} \right]. \quad (9)$$

The value of β is dominated, in most cases, by the term $(M_2 - M_1)/(2M_1)$. This means that the analyzer's sensitivity is mostly dependant on the contrast of the molecular weights between the gases in the mixture. As a consequence, an acoustical gas analyzer may not be able to distinguish one gas from another if they have nearly the same molecular weight. On the other hand, sonic gas analysis is an excellent tool for measuring the concentration of a relatively light gas in a heavier gas, or *vice versa*.

Notice in Eq. (8), that the effects of a varying temperature are now represented by the additive term $\delta T/[2T(0)]$. To eliminate these effects, this term must be subtracted from the relative frequency measurement.

For the purpose of illustration, this sensitivity analysis will be demonstrated here through the use molecular hydrogen gas H_2 as the contaminant (gas 2) in an atmosphere of air (gas 1). Both air and H_2 are primarily diatomic, so both have $\gamma \approx 7/5 = 1.40$, thus the polytropic coefficient can be ignored in the sensitivity analysis. Since the wavelength of the sound in the resonator is fixed by the resonator length, $\lambda = 2L_{eff} \approx 2L$, Eq. (1) shows that the sound speed and frequency of the fundamental plane wave resonance are directly proportional. The relative change in molecular weight $\delta M/M$ is related directly to the relative change in resonance frequency $\delta f/f(0)$:

$$\frac{\delta M}{M} = -\frac{2\delta f}{f(0)}. \quad (10)$$

The mean molecular mass of the binary gas mixture is the sum of their individual molecular masses ($M_{air} = 28.9644$ kg/kmole and $M_{H_2} = 2.01594$ kg/kmole) weighted by the concentration x of H_2 and the concentration $(1-x)$ of air.

$$\frac{\delta M}{M} = \left(\frac{M_{H_2} - M_{air}}{M_{air}} \right) x \approx -0.93x \Rightarrow \beta = \frac{\delta f}{f} \approx 0.465x \quad (11)$$

Electronic Phase Sensitivity

To determine how the sensitivity β relates to the minimum detectable contamination, one must address the specifics of the transducers (in particular, the phase-shifts between the loudspeaker's input current and sound pressure detected by the microphone, as a function of frequency) and of the electronic circuit chosen to track the resonance frequency as the concentration of the contaminant changes.

Several automated methods for determination of sound speed have been described. For example, an automated pulsatile time-of-flight system known as a "sing-around

velocimeter” was developed for measurement of sound speed in sea water [25]. A similar (but sinusoidal) self-resonant (positive feed-back) system was employed in a sonic gas analyzer designed to detect the presence of air in a helium gas recovery and re-liquefaction system [10].

Most sonic gas analyzers that have been constructed in my laboratory have used a phase-locked loop (PLL) to track the resonance frequency as the gas mixture concentration changes [14, 18, 19, and 21]. A PLL system, operating around 5 kHz, is used to track the resonance frequency in the system reported here. Such a PLL will have a typical phase stability of at least $\pm 0.1^\circ$ [26]. The rate-of-change of phase θ at the resonance frequency f_1 , with respect to frequency, is related to the quality factor Q_1 of the fundamental plane-wave resonance:

$$\left[\frac{d\theta}{df} \right]_{f_1} = \frac{360^\circ Q_1}{\pi f_1} = \frac{114.6^\circ}{f_1} Q_1. \quad (12)$$

For the resonance created with this apparatus in air, shown in Fig. 2, $(d\theta/df)_{f_1} = 1.44^\circ/\text{Hz}$. With the assumed phase stability of $\pm 0.1^\circ$, and Eq. (12) for the sensitivity β to an H_2 contaminate in air, the resolution of the resonator-PLL system is $\delta f/f = 12.5$ ppm. This corresponds to a concentration resolution of $\delta x \lesssim 2.5 \times 10^{-5} = 25$ ppm.

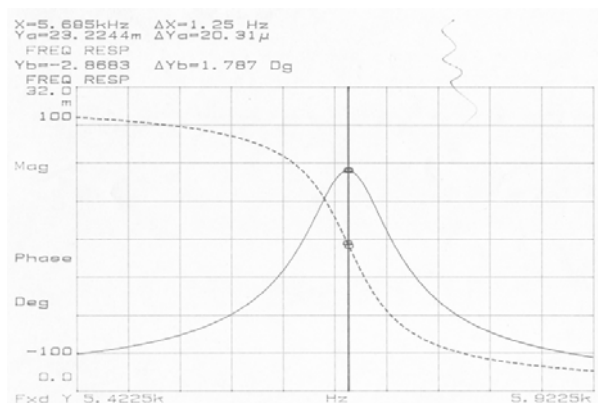


Figure 1. The magnitude (solid) and phase (dashed) of the frequency response of the resonator shown in Fig. 2, using two electrodynamic (moving-coil) transducers. The fundamental plane wave resonance frequency $f_1 = 5,685$ Hz and the quality factor, $Q = 71.6$.

Fan Noise Suppression

As shown in Fig. 4, the flow-through sonic gas analyzer also contains a fan that produces both broad-band flow noise and tonal noise related to the fan’s blade-passage frequency. Because of the intrinsic bandwidth-limiting provided by the high- Q resonator and the low-pass filter on the PLL’s mixer output, the fan noise is not a significant factor in the determination of the contaminant concentration sensitivity using this approach, even though the fan is quite close to the resonator.

With a speaker drive level of $0.5 V_{ac}$, the microphone output signal strength was $3.1 mV_{ac}$ at the resonance frequency f_1 . With no drive signal and no fan, the microphone signal was $0.3 \mu V_{ac}$ and with the fan and no drive signal, the microphone produced $2.0 \mu V_{ac}$. Therefore, the fan noise detected within the resonator at the resonance frequency was measured to be 64 dB below the signal level under these conditions.

Temperature Compensation

As can be seen in Eqs. (2) and (5) through (8), the mean absolute (Kelvin) gas temperature T plays a significant role in the determination of the sound speed. Although in most applications, the variation in mean gas temperature is relatively small, consideration of the mean temperature will be an important factor if the sonic gas analyzer will be able to discriminate between changes in temperature and the addition of a small concentration of a contaminant gas.

The flow-through sonic gas analyzer includes an electronic thermometer to compensate for the temperature dependence of the sound speed. The AD592 is a current-output precision integrated-circuit (IC) temperature transducer in a TO-92 (transistor) package [27]. The sensor generates a current of $1.0 \mu A/^\circ K$ (i.e., $293 \mu A_{dc}$ at $20^\circ C$). To maintain a concentration sensitivity of $\delta x/x = 25$ ppm, the temperature changes must also be detected to a relative precision of 25 ppm. In principle, a 16-bit A-to-D converter provides a resolution of 15 ppm. A dc-current meter in a typical data acquisition system has a thermal stability of $20 \text{ ppm}/^\circ C$ [28], so it appears that electronic temperature compensation based on the AD592 should preserve the predicted resolution *if* the gas mixture that enters the resonator is in thermal equilibrium with the copper resonator.

Thermal Equilibration Time

A detailed thermal analysis of thermal equilibration time requires specification of the air flow rate. Absent that measurement, a crude upper-bound can be placed on the thermal equilibration time constant using the geometry of the flow channel and the thermal properties of the air, copper, and PVC.

For the arrangement shown in Section 4, the inside diameter of the PVC pipe surrounding the copper end cap is $D = 26.06$ mm, while the outside diameter of the cap is approximately 24.54 mm. If the cap is co-axial with the pipe, then there is an annular gap $g = 0.76$ mm that is $\pi D = 8.2$ cm in circumference. The thermal equilibration time constant τ is the ratio of the heat capacity of the air $C = c_p \rho \pi D g L$ in the gap of axial depth $L \cong 1$ cm, to the thermal resistance R_{th} of the air in the annular gap of thickness g .

To estimate the maximum thermal resistance, we can assume laminar gas flow. In a duct of rectangular cross-section ($g \ll D$), the laminar flow Nusselt number [29], $N_D = 7.54 = D_h h / \kappa$, where $D_h = 2g = 1.52$ mm is the hydraulic diameter of the duct, $\kappa = 2.6 \times 10^{-2} \text{ W/m} \cdot ^\circ K$ is the thermal conductivity of air under standard conditions, and h is the convective heat transfer co-efficient. The thermal resistance of the air in the annulus is then $R_{th} = \pi D L h$.

$$\tau = \frac{C}{R_{th}} = \frac{g^2 \rho c_p}{3.77 \kappa} \quad (14)$$

As expected, this thermal time constant τ depends upon the square of the critical gap dimension g^2 and is independent of the pipe diameter D and flow length L , since both the thermal resistance R_{th} and the heat capacity C are proportional to those two larger dimensions. Substitution of the other thermophysical properties of air, $c_p = 1005 \text{ J/kg} \cdot ^\circ K$ and $\rho_{air} = 1.192 \text{ kg/m}^3$, into Eq. (14) yields a thermal equilibration time constant $\tau = 7$ msec.

The air flow will be laminar if the Reynolds number, $Re_D = \nu \rho D_H / \mu < 2,000$, where ν is the flow velocity within the annulus and $\mu = 1.83 \times 10^{-5}$ Pa-sec is shear viscosity of air. Substitution requires $\nu < 20$ m/sec for the flow to remain laminar, corresponding to a maximum volume flow rate $U < 1.2$ liters/sec, far greater than any expected flow rate.

Since the flow-through sonic gas analyzer uses the PVC fan housing to provide one of the gap surfaces and the resonator's copper body to provide the other, it is possible to calculate the ratio of "accessible" solid heat capacity to determine which surface dominates the air temperature. This can be quantified by forming the ratio of the "accessible" heat capacity per unit surface area which is the product of the thermal penetration depth $\delta_x = (2\kappa\tau/\rho c_p)^{1/2}$ and the heat capacity per unit volume ρc_p where now the values of the thermophysical constants are those of the two solids. The accessible heat capacity per unit area is therefore proportional to $[\kappa\rho c_p]^{1/2}$. For copper and PVC, that ratio is approximately twenty-seven, showing that the copper's temperature will dominate the air temperature in the gap.

Resonator and Gas Flow Manifold

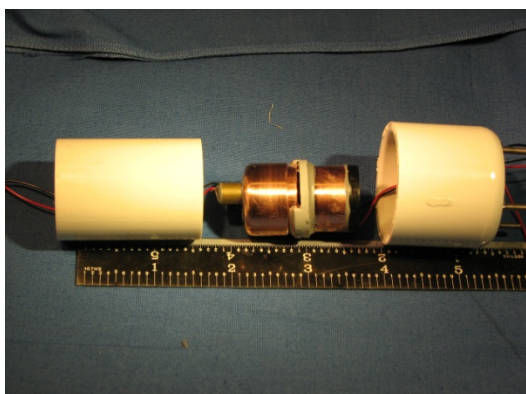


Figure 2. Photo of the resonator with the flow manifold separated exposing one mid-plane slit and the plastic ring that fits snugly in the PVC pipe, forcing the air to enter one slit and leave through the other slit (not visible), propelled by the fan in the PVC cap shown in Figure 4.

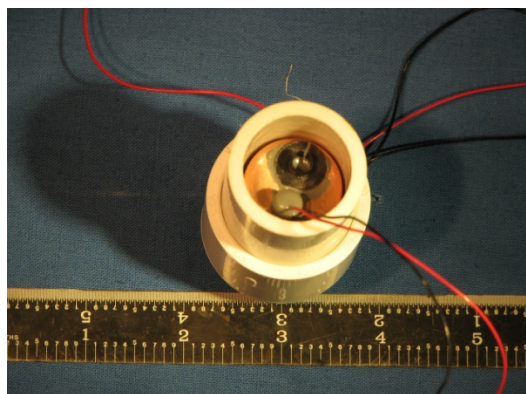


Figure 3. View of the flow-through sonic gas analyzer from the gas entry side. Air is sucked through the annulus created by the inside of the PVC pipe and the outside of the microphone cap. Attached to the cap is a heat sink fixture (TO-92) in excellent thermal contact with the copper resonator. The leads (red and black wires) from a Panasonic electret microphone are also visible. The heat sink holds the solid-state IC temperature sensor (AD 592).

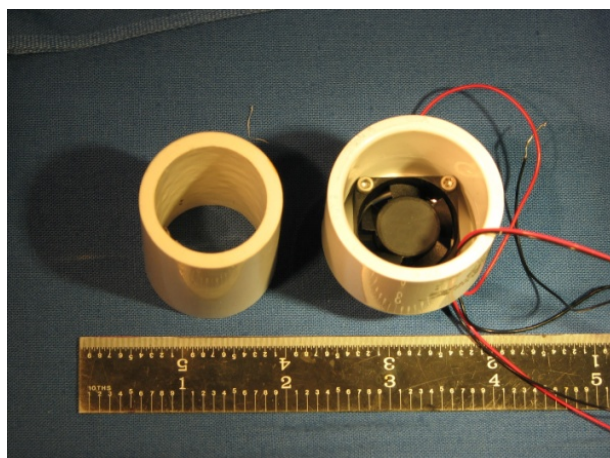


Figure 4. The 12 V_{dc} fan is mounted using three screws to the inside of the PVC cap. The fourth screw hole is used as a passage for the speaker and fan electrical leads.

Conclusion

Flow-through sonic gas analyzers might provide an attractive alternative to non-specific gas analyzers based on the measurement of thermal conductivity of gas mixtures since the concentration determined by the sonic method requires very little electrical power and does not require a reference cell. The concentration result is independent of the excitation amplitude, insensitive to the gas flow rate, can utilize electronic temperature compensation, and provides a digital (frequency) output that is well-suited to a multiplexed multi-sensor system.

Acknowledgments

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