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Measuring thermophysical properties of gases with a single thermocouple: Peltier vacuum gauge

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Abstract

Using a single thermocouple as a heater and temperature sensor simultaneously, we have developed a new type of gas pressure sensor termed the Peltier vacuum gauge (PVG). The PVG measures gas pressure by allowing an AC current carrying thermocouple, which is in contact with gases, to measure its own temperature. An AC current carrying thermocouple generates Joule heating along the wire and Peltier heating/cooling at the junction, and its temperature is determined by the thermophysical properties of surrounding gases which in turn depends on pressure. The complementary nature of the Peltier effect and the Joule heating allows the PVG to possess an excellent sensitivity over a wide range of pressure from 10^{-4} Torr to atmospheric pressure.

Keywords: Peltier tip calorimeter; Thermophysical properties of gases; Vacuum sensor

1. Introduction

A thermal conductivity vacuum gauge is a pressure measuring device in which the measured response is associated with energy loss from a heating element to surrounding gases [1]. The energy loss from the heating element of a gauge is due to thermal conduction through the surrounding gases, to the wire supports, and by radiation. Designating the rates of these energy losses by W_g, W_w, and W_r, respectively, we may write the total rate of heat transfer W_t as:

$$W_t = W_g + W_w + W_r \tag{1}$$

It is the first term which is dependent on pressure, and W_w and W_r constitute a constant background loss. In other words, the pressure dependence of the thermal conductivity of gases in the molecular flow regime is exploited in the vacuum gauge, and the magnitude and stability of the background determine the lowest measurable pressure. As the pressure level approaches the viscous regime where the mean free path of gases becomes short, the thermal conductivity of gases becomes pressure independent. In this regime, any increase (or decrease) of the number of molecules, as a function of pressure, participating in heat transport is compensated by the corresponding decrease (or increase) in the mean free path. Thus, the typical operating range of thermal conductivity gauges is from 10^{-3} to 1 Torr. Attempts to enhance the sensitivity of the gauges at the high pressure end have been made; examples of such attempts include moving the heating wire [2] and allowing gas convection [3].

Currently, two types of thermal conductivity gauges, the Pirani gauge and the thermocouple gauge, are in general use. Both of these gauges employ a heating wire and measure the temperature of the wire which is a function of pressure as a result of thermal conduction through surrounding gases. They differ only in the means of observing the wire temperature. The Pirani gauge measures the temperature of the hot wire from its resistance, and the thermocouple gauge uses a thermocouple attached to the wire. While thermal conductivity gauges are simple and sturdy, the major disadvantages of the gauges are their limited dynamic range and slow response time.

In this paper, we wish to introduce a new type of thermal conduction vacuum gauge utilizing the *Peltier effect* of a thermocouple in addition to the usual Joule heating. This new pressure sensor shall be called the Peltier vacuum gauge (PVG), and the basic idea behind the PVG is as follows: an AC current carrying thermocouple generates both Joule heating along the wire and Peltier heating/cooling at the junction, and the temperature

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of the thermocouple itself is determined by the thermophysical properties of surrounding gases which transport heat away from the thermocouple. Note that both heat capacity and thermal conductivity come into play in the AC method, while the DC method or steady state mode involves only thermal conductivity in the heat conduction. As shown in the next section, this then leads to the unique affirmative characteristic of the PVG, that is, its operating range covers from 10^{-4} Torr to atmospheric pressure.

2. AC heating method: line heater versus point heater

The purpose of this section is to present a heuristic argument, before expounding on the technical details of the PVG, which would clearly expose how the PVG achieves its sensitivity over such a wide pressure range. A thermocouple, a pair of dissimilar metallic wires with a junction, is the key element of the PVG. A thermocouple is normally used as a temperature sensor; in using a thermocouple as a temperature sensor, the Seebeck effect is utilized. On the other hand, the reverse effect, i.e., the Peltier effect, is also well known; when an electric current flows in a thermocouple, the junction acts either as a point heat source or a sink according to the current direction. It is emphasized that the PVG operates in the AC mode, that is, it measures gas pressure by flowing an AC current of a particular frequency through an thermocouple, which is in contact with gases, and then letting the thermocouple measure its own temperature. This method of using a single thermocouple as a heater and sensor simultaneously is an extension of the Peltier tip calorimeter which was previously developed in this laboratory [4].

Suppose a heat source supplies an oscillating power at angular frequency ω ($\equiv 2\pi f$ where f is the frequency) to a surrounding material. Then heat diffusion equation with thermal diffusivity D, which is equal to κ/C_p where κ denotes thermal conductivity and C_p does heat capacity per unit volume of the material, naturally provides a length scale λ:

$$\lambda = \sqrt{\frac{D}{\omega}} = \sqrt{\frac{\kappa}{\omega C_p}} \qquad (2)$$

λ is nothing but the distance within which the associated temperature oscillation decays, and from this thermal decay length one may define a probing volume Ω depending on the shape of the heater as follows:

$$\Omega \sim \begin{cases} \lambda^3 & \text{point source} \\ \lambda^2 L & \text{line source} \\ \lambda A & \text{plane heater} \end{cases} \qquad (3)$$

where L and A are the length and area of the line and plane heaters, respectively. Here, the finite size of the heaters is neglected for the sake of argument; this, however, may not be justifiable in real situations, in particular, for point sources.

For a given power the signal strength that the sensor (heater itself) detects, i.e., the amplitude of the temperature oscillation, would be inversely proportional to the total heat capacity of the probing volume Ω. Thus, the signal strength δT for the different heaters goes as:

$$\delta T \sim \frac{1}{C_p \Omega} \sim \begin{cases} C_p^{1/2} \kappa^{-3/2} & \text{point source} \\ \kappa^{-1} & \text{line source} \\ (C_p \kappa)^{-1/2} & \text{plane heater} \end{cases} \qquad (4)$$

Note that for line-type heaters only thermal conductivity appears in the above equation. Thus, thermal conduction sensors with line-type heaters would lose their sensitivity in the viscous regime even in the AC mode, because κ becomes pressure independent in the regime. On the other hand, for a point or plane heater the signal depends not only on κ but also on C_p, and C_p is proportional to pressure in the viscous regime. The PVG takes advantage of this point by utilizing the junction of a thermocouple as a point heat source arising from the Peltier effect. In reality, as already pointed out, the junction of a thermocouple is not a point source but it possesses a finite shape and is connected to the leads. Nevertheless, the final solution to the heat diffusion equation from a spherical source of finite size still involves heat capacity [5]. The PVG also makes use of the Joule heating effect to cover the low pressure region.

3. Principle of the Peltier vacuum gauge

Let us start with a situation illustrated in Fig. 1(a): a thermocouple consisting of two dissimilar metal wires, TC_L and TC_R, and a junction P_3 is placed in a vacuum chamber and connected to copper leads at P_1 and P_2. When an AC electric current oscillating at frequency ω, $I = I_0 \cos(\omega t)$, flows in the thermocouple, it generates two kinds of power: Joule heating $P_{JH} = I^2 R_0$ and Peltier heating/cooling $P_{PH} = \Pi I$, where R_0 and Π are the resistance and Peltier coefficient of the thermocouple, respectively. Π is related to the Seebeck coefficient ΔS of the thermocouple via temperature T as $\Pi = T\Delta S$. Note that P_{JH} occurs along the wire at frequency 2ω while P_{PH} is generated at the junction at ω. At this point two aspects of the PVG are emphasized again: first, the point-like dimensional characteristic of the junction combined with the AC method implies that the Peltier effect of a thermocouple would provide sensitivity in the high pressure region to complement the usual Joule heating effect of thermal conductivity sensors. Secondly, the AC method also provides a means of separating these effects, that is, they occur at different frequencies and consequently can be detected separately with a lock-in amplifier.

Now if the voltage appearing between P_1 and P_2 in the presence of an AC current is measured, it may be expressed as:

$$V = I_0 R_0 \cos \omega t + V_S(\omega) + V_S(2\omega) + V_R(\omega) + V_R(3\omega). \qquad (5)$$

The first term, of course, is a simple Ohmic voltage drop across the thermocouple caused by the current. The second and third terms represent Seebeck voltages which are related to the temperature oscillations δT_ω at ω and $\delta T_{2\omega}$ at 2ω, respectively, at the junction (P_3) via the Seebeck coefficient ΔS. δT_ω is induced by the power oscillation at the junction due to the Peltier effect and $\delta T_{2\omega}$ is caused by Joule heating along the wire; the amplitudes are determined by heat conduction from surrounding gases and thus carry information on pressure. Note that while heat is gen-

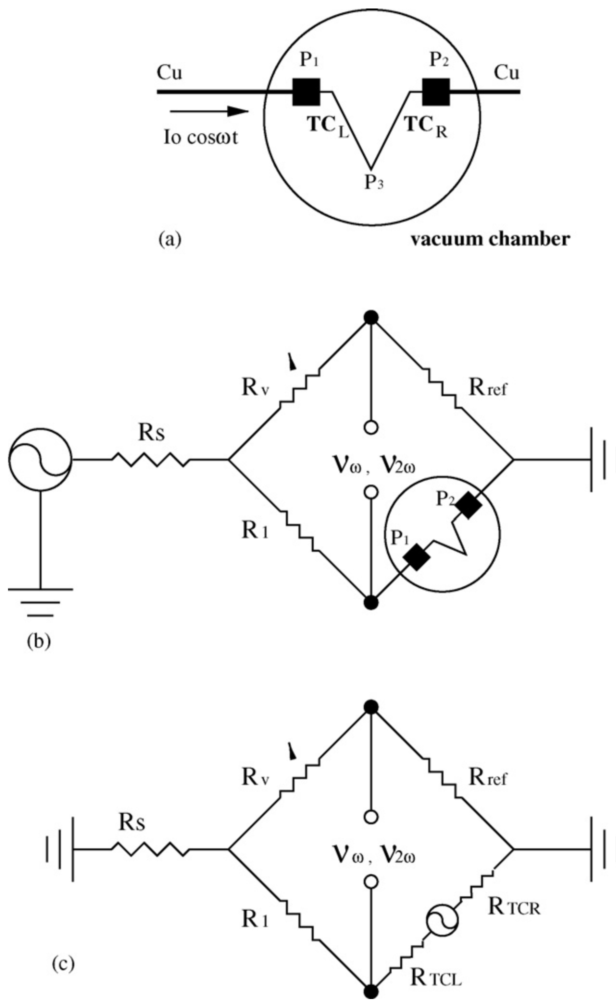

Fig. 1. Schematic diagram and operation principle of the Peltier vacuum gauge. (a) The voltage appearing between P_1 and P_2, when an AC electric current flows in the thermocouple, includes the Seebeck voltages caused by the Peltier effect at the junction and Joule heating along the wire. These Seebeck voltages carry thermophysical information of the surrounding gases and therefore the vacuum state of the chamber. TC_L, TC_R, and Cu denote two thermocouple wires and copper leads, respectively. The diameter of the thermocouple wires are either 25 or 12.5 μm, while that of the copper leads is 2 mm. (b) The measuring circuit adopting a Wheatstone bridge with a variable resistor R_v for balancing. R_s is the source resistance of 1 kΩ, and R_1 and R_{ref} are the bridge resistors with fixed values. The signals at ω and 2ω, V_ω and $V_{2\omega}$, are picked up with a lock-in amplifier. (c) In a symmetrical bridge a replica thermocouple is used as the reference resistor R_{ref}, and its junction is thermally grounded. The equivalent circuit for the measurement of the signals generated at the junction is indicated in the figure. All other signals except the Seebeck voltages from the junction disappear. R_{TCL} and R_{TCR} represent the resistance of thermocouple wires, and their sum is R_0.

erated by both the Peltier effect at the junction (at ω) and the Joule heating along the wires (at 2ω), there is no interference between these since the governing equation is linear. It may be noted that while the Peltier effect also occurs at the junctions P_1 and P_2 where thermocouple wires and copper leads meet, the associated temperature oscillations can be kept negligible compared to that at P_3 by making the junctions thick. For instance, thermocouple wires of diameter 25 or 12.5 μm may be used and copper wires of thickness 2 mm are attached to the thermocouple wires as leads.

In addition to the Seebeck voltage at 2ω, $V_S(2\omega)$, Joule heating along the wire also produces the fourth and fifth terms of Eq. (5) at frequencies ω and 3ω, respectively. As a result of self-heating the resistance of the wires would possess a small oscillating component at 2ω because the temperature coefficient of resistance of the thermocouple wires is not zero. Then the resistive voltage drop between P_1 and P_2, which is a product of the resistance and current, include $V_R(\omega)$ and $V_R(3\omega)$. Since the resistive heating is proportional to I^2, both $V_R(\omega)$ and $V_R(3\omega)$ are proportional to I^3. The well-known 3ω method is based on measuring the third harmonic signal $V_R(3\omega)$ from a metallic heater in contact with a sample [6].

The PVG we propose here is based on the fact that if one can isolate the Seebeck voltage $V_S(\omega)$, the second term in Eq. (5), out of the total signal between P_1 and P_2, one would be able to measure the pressure of the chamber in the viscous regime. The PVG, of course, is also used as a thermal conductivity sensor covering the low pressure region by simultaneously detecting the Joule heating signal $V_S(2\omega)$. Thus, the Seebeck voltages $V_S(\omega)$ and $V_S(2\omega)$ play complementary roles in the PVG and we shall call them the Peltier and Joule signals, respectively.

An obvious difficulty in isolating $V_S(\omega)$ is the presence of other contributions at the same frequency ω, that is, the first term and $V_R(\omega)$ in Eq. (5). It is rather straightforward to eliminate the first term, and this is achieved by adopting a Wheatstone bridge circuit as illustrated in Fig. 1(b). The IR_0 term is easily removed by balancing the bridge. However, even after balancing the bridge $V_R(\omega)$ still has to be eliminated from the measured signal ν_ω at ω to extract $V_S(\omega)$. This can be done by employing a symmetrical Wheatstone bridge with a replica of the PVG, a thermocouple of the same materials and of the same length and diameter, as the reference resistor. With $R_{ref} = R_0$, all unwanted signals would be canceled. In this case, however, it is important to thermally ground the junction of the reference thermocouple to a massive copper block to suppress the generation of Seebeck voltages from the reference arm. Then $\nu_\omega = V_S(\omega)$. Also the measured voltage $\nu_{2\omega}$ at 2ω is equal to $V_S(2\omega)$ for the Joule heating signal.

Although the symmetrical bridge is naturally balanced by itself, we continued to keep the variable resistor R_v as one arm of the bridge to allow a fine tuning. When the perfect balancing is achieved, the junction of the thermocouple in the chamber can be thought of as a signal source and the voltage signals appearing at frequencies ω and 2ω are detected with a lock-in amplifier. Fig. 1(c) is the equivalent circuit for measuring these signals $V_S(\omega)$ and $V_S(2\omega)$. The typical resistances for the Wheatstone bridge with K-type thermocouples (alumel–chromel) were R_1, $R_v = 100\ \Omega$, and R_0, $R_{ref} = 30\ \Omega$.

4. Performance of the PVG

In verifying the performance of the PVG, we first focused on the Peltier signal because the Joule heating effect shows similar behaviors to that in conventional thermal conductivity sensors in the DC mode. K-type (alumel–chromel) thermocouples were used, and the input current amplitude was kept low to contain the amplitude of temperature oscillation less than 1 K. At this power level it turned out that the Joule heating effect ($\sim I^2$) is much smaller than the Peltier effect ($\sim I$). A lock-in amplifier (EG&G DSP7260) with a built-in synthesized signal source was used for data acquisition. In the latter part of this section we will illustrate the Joule heating effect on the sensitivity of the PVG by increasing power to the level where the Peltier and Joule heating signals are comparable.

Let us start with a simple situation, that is, a thermocouple sitting alone in a low pressure chamber ($\leq 10^{-5}$ Torr). As can be seen in Fig. 1(a), the heat generated at the junction point P_3 by the Peltier effect would diffuse away to the thermal bath via conduction through the thermocouple wires. At this pressure level gaseous conduction is small compared to wire conduction. Since the length and diameter of the two thermocouple wires, TC_L and TC_R, are equivalent, the situation may be simplified to the case of a single wire with the same length and diameter, thermal conductivity $\kappa^{\text{wire}}(\equiv \kappa^{TC_L} + \kappa^{TC_R})$, and heat capacity (per unit volume) $C_p^{\text{wire}}(\equiv C_p^{TC_L} + C_p^{TC_R})$ connected to the heat bath. As a sinusoidal power $P_0 \exp(i\omega t)$ is generated at the junction point P_3, the temperature oscillation at the same point is given by solving the one-dimensional heat diffusion equation:

$$\delta T_\omega = \frac{j_0}{\kappa^{\text{wire}} k^{\text{wire}}} \tanh(k^{\text{wire}} d), \tag{6}$$

where d is the length of the wire, k^{wire} the thermal wavenumber defined as $k^{\text{wire}} = \sqrt{i\omega C_p^{\text{wire}}/\kappa^{\text{wire}}}$, and j_0 is the power density, i.e., P_0 divided by the cross-section area of the wire.

Fig. 2 is the plot of δT_ω as a function of frequency. The amplitude of the current applied was 0.225 mA (rms). The real and imaginary parts of δT_ω were converted from the measured values of v_ω using the Seebeck coefficient of the thermocouple. The diameter of the thermocouples were either 25 or 12.5 μm; the length was fixed at d = 2.4 mm. As can be seen from Eq. (6), the thermal conductivity of the wire is easily obtained from the DC limit, $\delta T_\omega = (j_0 d/\kappa^{\text{wire}})$, while the thermal wave number (and therefore heat capacity) of the wire can be determined by fitting the whole data set, both real and imaginary parts, to Eq. (6). The fitting lines in the figure indicate that Eq. (6) describes the situation well. The curve fitting yielded the values of κ^{wire} and C_p^{wire} at room temperature to be 49.3 W/m K and 8.40 J/cm^3 K, respectively. These values are in perfect agreement with those of literature [7].

Now we turn to the pressure region where the gaseous conduction is dominant. Here, δT_ω would become pressure dependent, and the junction temperature was measured as a function of pressure. Dry nitrogen gas was used as a test gas. A spinning rotor gauge (SRG) was used as the reference standard gauge in the pressure range between 10^{-4} and 10^{-1} Torr, while two capacitance diaphragm gauges (CDG) were adopted between 10^{-2} Torr and atmospheric pressure. In setting up the vacuum system and gas admission system, we followed the direction of *recommended practice for calibrating vacuum gauges of the thermal conductivity type* by the Thermal Conductivity Gauging Committee of the American Vacuum Society [8]. Fig. 3 illustrates the pressure-dependent δT_ω measured from 10^{-3} Torr to atmospheric pressure as a function of frequency. It is seen from the figure that the magnitude of the real and imaginary parts at various frequencies decreases as pressure increases. In particular, the magnitude of the imaginary part seems to be sensitive to a pressure change at high pressures.

In order to reveal its pressure dependence more clearly, we plotted the normalized signal at various frequencies as a function of pressure in Fig. 4, that is, at each frequency the value of $|\text{Im}(\delta T_\omega)|$ as a function of pressure was normalized against its value at the low pressure limit. The striking feature in Fig. 4 is evident, that is, as frequency increases, the pressure-sensitive region moves to higher pressures. Thus, the Peltier signal shows appreciable sensitivity even in the viscous regime when the PVG is operated at, say, 10 Hz. This conspicuous result is obviously due to the fact that the PVG is driven in AC mode and the Peltier heat source is a point-like one. To understand the frequency-dependent sensitivity of the PVG, consider a point heater with a radius embedded in a fluid. If a sinusoidal power at frequency ω with density j_q is applied to the heater, δT_ω at $r = a$ is obtained

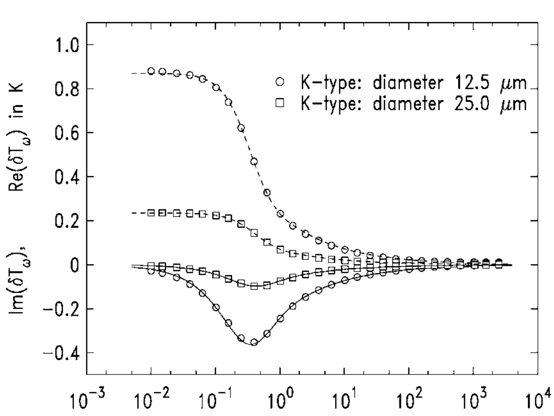

Fig. 2. The real and imaginary parts of δT_ω, obtained for a thermocouple in a vacuum below 10^{-5} Torr, is plotted as a function of frequency. K-type thermocouples with diameter 25 and 12.5 μm were used. The symbols denote the data points, and the dotted (real parts) and solid (imaginary parts) lines represent the fitting results. The measured data were fitted to Eq. (6).

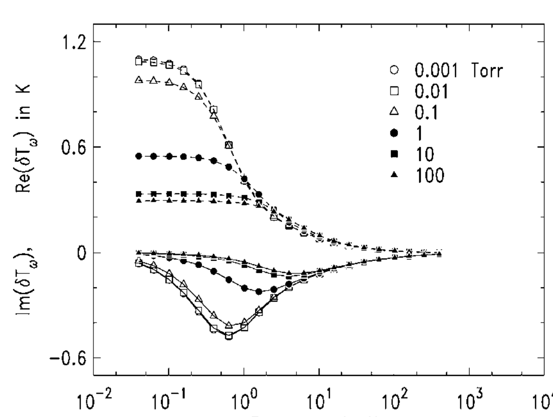

Fig. 3. Frequency dependence of real (dotted lines) and imaginary parts (solid lines) of δT_ω from the PVG at various pressures. We used a thermocouple of diameter 25 μm and total length 1 mm. For the suppression of Joule heating effects such as $V_S(2\omega)$, $V_R(\omega)$, and $V_R(3\omega)$ in Eq. (5), $|\delta T_\omega|$ was kept less than 1 K.

by solving the heat diffusion equation [5]

$$\delta T_\omega = \frac{j_q a}{\kappa_s}\frac{1}{1+ak_s} \tag{7}$$

with k_s and κ_s denoting the thermal wavenumber and conductivity of the surrounding fluid, respectively. The effect of the finite mass of the point heater was ignored in deriving Eq. (7). As can be seen from the equation, not only κ_s but also $k_s\left(\equiv\sqrt{i\omega C_{ps}/\kappa_s}\right)$ is involved in δT_ω, while only κ_s would appear in the DC mode. For rigorous account of the phenomenon, of course, one must take the fact that the heat generated at the junction also leaks out through the wires into consideration; this can be done by superposing Eqs. (7) and (6) in an appropriate way as was done in Ref. [4]. For the present purpose, however, a qualitative understanding would suffice and we may safely conclude that the pressure dependence of the heat capacity results in the frequency-dependent sensitivity of Fig. 4.

It is particularly significant for the performance of the PVG to establish that the Peltier signal maintains its sensitivity at high pressures or in the viscous regime. Having done so, we carried out simultaneous measurements of the Peltier and Joule signals at ω and 2ω, respectively. In order to make the two signals comparable, the input current was increased to 1 mA (rms) which is about five times as large as the one used in the above measurements. At this current level the amplitude of the temperature oscillation was still less than 5 K. Fig. 5(a) is the plot of the normalized Peltier and Joule signals as a function of pressure from 10^{-4} Torr to atmospheric pressure. The measuring frequency was fixed at 20 Hz. It is seen from the figure that the Joule heating signal covers the low pressure side, while the Peltier signal is sensitive in the high pressure region. It is also worth noting that the Joule signal shows a reasonable sensitivity even below 10^{-3} Torr. This is probably because the AC method is more sensitive to tiny signals than the DC method. Fig. 5(b) displays the sensitivity of the two signals as a function of pressure; the sensitivity here is defined as the derivative of the normalized signal with respect to the logarithmic pressure. Thus, the complementary nature of these two signals allows the PVG to

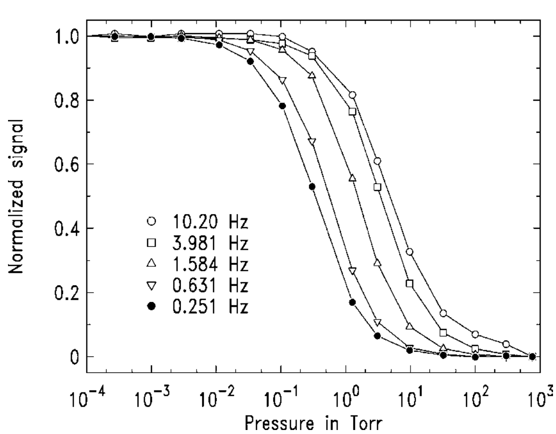

Fig. 4. Normalized Peltier signals as a function of pressure. At each frequency the value of $|\mathrm{Im}(\delta T_\omega)|$ as a function of pressure was normalized against its value at the low pressure limit. Note that the pressure-sensitive region moves to higher pressures as frequency increases.

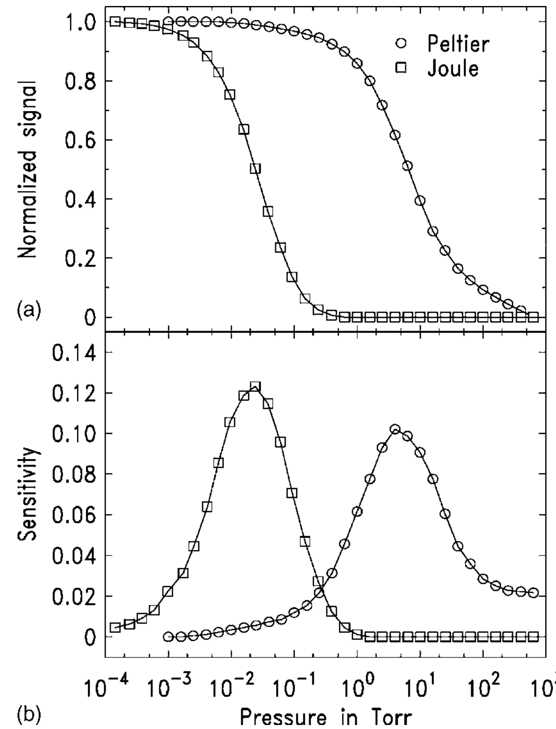

Fig. 5. Normalized Peltier and Joule signals measured at 20 Hz. (a) The plot of the normalized signals as a function of pressure from 10^{-4} Torr to atmospheric pressure. The Joule heating signal covers the low pressure side, while the Peltier signal is sensitive in the high pressure region. (b) The sensitivity of the two signals as a function of pressure. The sensitivity is defined as the derivative of the normalized signal with respect to the logarithmic pressure.

possess a distinctive characteristic as a vacuum sensor: an excellent sensitivity over a wide range of pressure from 10^{-4} Torr to atmospheric pressure.

5. Summary

In summary, we have developed a new type of gas pressure sensor, termed the Peltier vacuum gauge, using a single thermocouple as a heater and temperature sensor simultaneously. The PVG measures gas pressure by allowing an AC current carrying thermocouple, which is in contact with gases, to measure its own temperature. A current carrying thermocouple generates Joule heating along the wire and Peltier heating/cooling at the junction, and its temperature is determined by the thermophysical properties of surrounding gases which in turn depends on pressure. The PVG possesses distinguished features as a pressure sensor: first, it has an excellent sensitivity over a wide range of pressure from 10^{-4} Torr to atmospheric pressure. Secondly, its simple structure offers a way for miniaturization of the vacuum sensor appropriate for use as a local and fast probe, and finally it operates only several degrees above the surrounding temperature.

Acknowledgments

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