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## A NEW FERROELECTRIC TRANSDUCER FOR USE IN HEAT TRANSFER AND FLOW STUDIES

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ABSTRACT

A transducer is described for the measurement of heat transfer rates in gaseous systems. Based on the ferroelectric properties of tri-glycine sulphate ( $(\text{CH}_2\text{NH}_2\text{COOH})_3 \cdot \text{H}_2\text{SO}_4$ ), the device autostabilizes its operating temperature so that external heat-transfer depends linearly upon the ambient temperature and the film coefficient. An analysis of temperature stability in several heat-sensitive transducers is presented, and the fundamental improvement in stability offered by the ferroelectric probe is demonstrated both in the analysis and experimentally. Data are presented in reference to possible applications of the device as a linear heat-transfer gauge and a binary gas analyzer. A qualitative discussion of transient response is given, and a comparison is made to a similar technique for the linear detection of diffusion-controlled processes in convected mass-transfer systems.

INTRODUCTION

This paper reports on experimentation related to the development of a novel transducer which has applications to heat transfer studies, fluid dynamics, temperature and pressure detection, and a variety of similar processes requiring sensors which react to heat flux. Although several well-known techniques are already available for such measurements, the method described here offers a new dimension of stability while requiring no more, and perhaps ultimately fewer, complicated control mechanisms. The question of stability is of great importance in relation to the utility of the device for unsteady measurements while the prospect of a simpler instrument package offers obvious advantages in the actual operation. A general description of the technique will be given here, and a more detailed analysis of the crucial physical characteristics of the transducer will be presented in the next section.

The method is based upon a heat-sensitive element utilizing the non-linear dielectric properties of crystalline tri-glycine sulphate. This material undergoes a second-order phase transition from the ferroelectric to the paraelectric

state at its Curie temperature ( $49.4^\circ\text{C}$ ) and exhibits pronounced dielectric non-linearities in the vicinity of this temperature. A single crystal of tri-glycine sulphate (TGS) is cut into a thin wafer with its plane-parallel faces perpendicular to the ferroelectric, monoclinic  $b$  axis. Silver or gold electrodes are evaporated onto these faces and a tiny lead wire attached to each electrode. The assembly is then installed in a probe for connection to external circuitry and exposure to the system under study.

When the crystal, thus fitted, is placed in contact with a quiescent or flowing gas and heated dielectrically through the application of an alternating voltage, its temperature rises until it undergoes a rather abrupt transition into an "autostabilized" state. In the neighborhood of this state the internal heat generation of the crystal falls rapidly with increasing temperature while the external heat transfer from its surface, as one would expect, rises in direct proportion to the ambient conditions. A stable operating point results when the internal and external rates of heat flow become equal. When the ambient conditions change due to variations in velocity, thermal conductivity of the gas, pressure or temperature, a new operating point is automatically established such that the actual surface temperature of the element changes by only a small amount. As long as the ambient temperature remains sufficiently below the Curie temperature (more than about  $10^\circ\text{C}$ ) and the applied voltage is held above some minimum value, the TGS crystal will remain in the autostabilized condition, constantly adjusting its temperature by small increments to maintain the thermal steady state. Inasmuch as the rate of heat transfer from the element is always equal to the rate of heat generation from dielectric heating, the magnitude of the heat flow can always be determined through an appropriate measurement of impedance, voltage across the crystal and/or current through it. Briefly, then, the transducer functions as a "linear" heat-transfer gauge and may be used for absolute measurements of heat flow or to detect changes in any property of a system that affects energy exchange between the system and the autostabilized crystal.

In addition to several fundamental investigations of the ferroelectric behavior of tri-glycine sulphate and its crystalline isomorphs tri-glycine selenate and tri-glycine fluoberyllate (6,9), a number of articles have appeared

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recently (4,5,7) which suggest some applications in addition to those discussed in this paper and which also attempt to explain certain other phenomena associated with the temperature-autostabilized regime. In keeping with the practice of other authors who have worked with these devices, the acronym TANDEL (Temperature Autostabilized Non-Linear Dielectric Element) will be adopted here.

#### THE MECHANISM OF TEMPERATURE STABILITY IN HEAT-SENSITIVE PROBES

##### A Classical Example

Any device which can be used to measure rates of heat flow, or any of the thermodynamic or transport properties of a system that influence heat flow, must, in the final analysis, function so as to establish a thermal steady state between itself and the system under measurement. Perhaps the best example of this is the classical wet-bulb thermometer. This simple device consists of a conventional liquid-in-glass thermometer with the bulb covered by a wick saturated with a fairly volatile liquid. When the device is exposed to an atmosphere containing a mixture of non-condensable gases with the vapor of this liquid, a steady state is set up in which the temperature indicated by the thermometer is appreciably lower than the actual ambient temperature. In reality, the steady state simply reflects a balance between the amount of sensible heat transfer to the wick-bulb system from the warmer ambient and the amount of latent heat transfer in the opposite direction due to evaporation of liquid into the (presumed) unsaturated gas. From this temperature reading one can determine any single thermodynamic or transport property of the system if all other such properties that appear in the heat-balance equation are known. A change in any one of these variables will result in the establishment of a new steady state and a new temperature reading. Therefore, with a little experimental skill (and patience), one could actually use such a device as a heat-transfer gauge and determine the influence of changes in ambient temperature, convection level, or any other relevant variable upon the energy exchange process.

Aside from the obvious drawbacks associated with such a cumbersome technique, the most serious disadvantage to this "transducer" is that it is highly non-linear. If one is really interested in determining changes in, say, the overall heat transfer coefficient  $h$  from measured changes in heat flow  $q$ , one would like to be able to use a simple linear relation of the form

$$q = h(T_a - T_s) \quad (1)$$

where  $T_a$  and  $T_s$  are the ambient and transducer-surface temperatures, respectively. Clearly, since the wet-bulb thermometer changes its surface temperature  $T_s$  at each new steady state, equation (1) doesn't offer the desired linearity. In fact, the best that one could hope for would

be some kind of calibration curve, probably specific for a given set of experimental conditions.

##### Conventional Stability

Now let us consider the family of heated-element probes, examples of which are the hot-wire sensor, the bead thermistor sensor, and the ferroelectric transducer under discussion here. In each case, just as in the elementary one discussed above, the analysis points to the establishment of a thermal steady state between the probe and the medium in which it is placed. The obvious advantage offered by these devices relates to their being heated electrically, thereby making possible continuous monitoring of the heat flow with appropriate instrumentation. Even in view of this experimental simplification, however, the linearity of equation (1) remains a requirement if we are to infer variations in one variable strictly from measured variations in any other single variable. It seems reasonable, then, to consider the mechanisms whereby each of these sensors tends to stabilize its "operating" temperature so as to linearize equation (1) to a greater or lesser extent.

The word "operating" is set aside here since the thermal steady states set up by these devices are not thermodynamic equilibrium states. That is, temperature gradients exist within the body of the probe itself as well as in the medium being studied, and the surface temperature is only a measure of the actual temperature distribution inside. But since the driving force for external heat transfer depends upon the surface temperature and not upon the distribution, stability of this variable appears to be the appropriate criterion to examine in deducing the linearity of (1) with a particular technique.

In the case of the hot wire, the expression for the heat generation due to excitation by an ac heating voltage with an effective value of  $E$  volts rms is

$$q = \frac{E^2}{R_0[1 + \alpha(T_s - T_0)]} \quad (2)$$

in which the temperature dependency of the wire resistance is assumed to be that associated with a normal ohmic device. That is, the sensor has a resistance of  $R_0$  ohms at temperature  $T_0$  and a positive temperature coefficient of resistance of  $\alpha R_0$  ohms/degree. The thermal steady state which will be set up between the hot wire and its surroundings may be characterized, then, by setting equations (1) and (2) equal to each other (with, of course, the gradient in equation (1) reversed). This accounts for all possible sets of operating conditions for which the heat generated within the probe is precisely equal to the heat transferred from it.

One can now inquire as to what will be the temperature stability of the hot wire. In other words, how will  $T_s$  change in response to small changes in  $h$  or  $T_a$  in equation (1), thereby influencing the linearity of the  $q$  dependence. Although we can determine this for changes in

either of these variables, it is perhaps simpler to examine the case in which  $T_a$  changes and defer the  $h$ -varying case to a separate discussion. One, therefore, defines a stability  $S$  in the following manner:

$$S = \left( \frac{\partial T_s}{\partial T_a} \right) E, h \quad (3)$$

where  $T_s$  is obtained in terms of the other variables from the equated relations (1) and (2). The result of this differentiation is

$$S_{\text{hot wire}} = \frac{1}{2} + \frac{(T_a + \frac{1}{\alpha} - T_0)}{2 \sqrt{(T_a + \frac{1}{\alpha} - T_0)^2 + \frac{4E^2}{h\alpha R_0}}} \quad (4)$$

Two important characteristics of this result are immediately apparent. First, the hot wire stability approaches unity at large values of  $T_a$  (with, of course,  $T_s$  remaining greater than  $T_a$  in all cases). Secondly, since the term  $(T_a + \frac{1}{\alpha} - T_0)$  is positive for all reasonable conditions, the stability varies between 1/2 and unity. It follows, then, that changes in the ambient temperature around a hot wire sensor result, at steady-state, in changes in the probe temperature at least half as large. This stability obviously depends upon the effective heat-transfer coefficient  $h$ , but insofar as the parenthesized term in equation (4) remains positive,  $S$  remains above 1/2.

The case of the thermistor sensor can be treated similarly. The analogous heat generation expression is

$$q = \frac{E^2}{R_0} e^{\beta \left( \frac{1}{T_0} - \frac{1}{T_s} \right)} \quad (5)$$

where the usual exponential dependence of thermistor resistance upon temperature is assumed, and  $\beta$  is a positive "material" constant. Equating the generation and loss terms (equations (5) and (1)), solving for the thermistor temperature  $T_s$ , and differentiating to obtain the stability, one arrives at the following result:

$$S_{\text{thermistor}} = \left[ 1 - \frac{\beta(T_s - T_a)}{T_s^2} \right]^{-1} = \left[ 1 - \frac{\beta q h}{(q + h T_a)^2} \right]^{-1} \quad (6)$$

Since for all reasonable operating conditions  $T_s < \beta/2$ , equation (6) approaches unity only in the limit of very low heat generation and correspondingly low ambient temperatures. Secondly, since the heat-transfer coefficient must always be greater than  $q\beta/T_s^2$ , one observes that the stability is always greater than this limiting value. Physically, this means that changes in the ambient temperature around a thermistor probe result, at steady state, in changes in  $T_s$  at least as large. Thus, where problems of stability are important in relation to the linearity of equation (1), the thermistor sensor (clearly because of its negative temperature coefficient of resistance) is inferior to the hot wire.

It should be emphasized that transient response is not involved in this analysis. The

changes envisioned here are between pairs of operating points, each characterized by the equivalence of heat generation within and transfer from the probe. To extend this reasoning so as to include transient behavior as well, one must then consider the time dependent processes occurring both inside the sensor and in the system under study.

#### The Autostability of TGS-TANDEL

The mechanism by which a crystal of ferroelectric tri-glycine sulphate (prepared as in the previous description) "autostabilizes" its operating temperature may be understood through an examination of the variables which influence heat generation inside the crystal due to dielectric losses in the ac electric field. For a crystal of volume  $V$  subjected to a sinusoidal electric field of amplitude  $E_0$  volts per unit length in the direction of its ferroelectric axis, the rate of heat generation is given by

$$q = \frac{\omega V \epsilon'' E_0^2}{2} \quad (7)$$

where  $\omega$  is the frequency of the field, and  $\epsilon''$  is the imaginary part of the complex permittivity measured, also, along this axis. It should be noted that  $\epsilon''$ , itself, depends upon frequency, field strength and crystal temperature, and indeed, it is the nature of this dependence that makes possible the phenomenon of autostabilization.

If one visualizes, as in the previous two stability analyses, a TGS TANDEL probe exposed to some gaseous system at a temperature  $T_a$ , the effect of increasing the voltage across the crystal will be to increase the rate of heat generation, thereby increasing the temperature of the succession of thermal steady states through which the total system passes. Due to the increase of  $\epsilon''$  with temperature, a critical voltage is reached, above which the heat generation exceeds the heat loss, and the crystal temperature rises rapidly. This transition point occurs when the following two conditions are met simultaneously:

$$\frac{\omega V \epsilon'' E_0^2}{2} = h(T_s - T_a) \quad (8)$$

$$\left. \frac{\partial \epsilon''}{\partial T} \right|_{\omega, E_0, T_s} = \frac{2}{\omega V E_0^2} \quad (9)$$

where it is important to realize that equation (8) holds for all steady states while equation (9) applies only to the final state before the transition.

In the vicinity of the Curie point  $\epsilon''$  reaches its maximum value at a temperature  $T_m$  and then falls rapidly with temperature in the region just above  $T_m$ . When the transition described above occurs, the crystal spontaneously changes its operating state so as again to satisfy equation (8) at a temperature  $T_s > T_m$ . The significant difference between this new state and those prior to the transition

arises because of the rapid change of  $q$  with  $T_S$  due to the abrupt decrease of  $\epsilon''$  in this temperature region just above  $T_M$ . An increase in the crystal voltage at this point results in only a very small rise in  $T_S$  and, therefore, a correspondingly small variation in  $q$ . To the extent to which the decrease in  $\epsilon''$  counteracts the increase in  $E_0^2$  for such a change, the left side of equation (8) remains invariant.

Decreasing the voltage below the critical value that initiated the transition does not immediately result in a return to the low-temperature state. For conditions of constant  $h$  and  $T_A$  (which we have tacitly assumed throughout this analysis), the dependence of  $\epsilon''$  on both crystal temperature and applied voltage is such that the high-temperature steady states prevail over a significant range of decreasing voltage. When at last, for some low value of  $E_0$ , a steady state can no longer be found on that portion of the relevant  $\epsilon''$  curve above  $T_M$ , the crystal returns to the low-temperature state. This behavior establishes a temperature-voltage hysteresis loop with the high-temperature state being that of autostabilization. Data for a typical crystal heated at a frequency of 100 kHz in an atmosphere of carbon dioxide maintained at  $T_A = 31.1^\circ\text{C}$  are presented in Figure 1.

Having demonstrated the existence of the autostabilized state, one can now compute the stability to be expected with TGS TANDEL. That is, when the ambient temperature (again using  $T_A$  as the variable) changes by  $\Delta T_A$ , thus establishing a new steady state, what will be the change in the crystal temperature  $\Delta T_S$ .

Dvořák et al (3) have obtained a rigorous expression for the stability of the ferroelectric sensor through reasoning similar to that presented above for purely resistive probes. They find

$$S_{\text{TANDEL}} = \frac{\partial T_S}{\partial T_A} \Big|_{\omega, E_0, h} = \left[ 1 - \frac{(T_S - T_A)}{\epsilon''} \frac{\partial \epsilon''}{\partial T} \Big|_{T=T_S} \right]^{-1} \quad (10)$$

Equation (10) shows that the stability lies between zero and one as long as  $\epsilon''$  is a decreasing function of temperature. Substitution of data obtained at a frequency of 20 kHz for  $\epsilon''$  and its temperature derivative led these authors to predict stability values on the order of 0.1 to 0.2. Experimental stabilities determined by these workers and also by the present authors have supported this result and have in some cases dropped significantly below 0.1.

It may be concluded, therefore, that the autostabilized ferroelectric transducer exhibits a temperature stability on the order of one fifth that shown by a hot wire probe and on the order of one-tenth that of a thermistor. Although one is limited with a particular TANDEL to applications wherein the ambient temperature is at least about ten degrees below the Curie temperature of the ferroelectric material, there

are compounds other than tri-glycine sulphate that exhibit this behavior, and some of these have Curie temperatures above  $50^\circ\text{C}$ . Techniques have also been described (1) to obtain mixed crystals having Curie points intermediate to those of the pure components.

#### EXPERIMENTAL PROCEDURES AND RESULTS

The experimentation conducted thus far in this laboratory with the TGS-TANDEL transducer has dealt with the following problems:

1. Growth of raw material and fabrication of this into several suitable probes.
2. Development of the instrumentation needed to study the device under conditions of interest.
3. Confirmation of the existence of the autostabilized state through the observation of temperature-voltage hysteresis curves.
4. Demonstration of the utility of the device as a linear heat-transfer gauge and also as a binary gas analyzer in conjunction with experimentally determined calibration curves.
5. Qualitative observations of the response of the transducer to unsteady conditions in the gas surrounding it.

#### Crystal Preparation

Tri-glycine sulphate single crystals were grown out of a saturated aqueous solution of glycine and sulphuric acid. The crystal has a cleavage plane perpendicular to the ferroelectric axis, and after confirmatory X-ray analysis, sample orientation was based upon the cleavage faces. After grinding, etching and polishing, according to a technique developed by Chynoweth and Abel (2), the resulting wafers had dimensions on the order of 1 mm thick and 2-4 mm in diameter. Gold or silver electrodes were then vacuum deposited on the parallel faces, and 0.003" platinum lead wires were attached to the electrodes with conducting cement. On a few of the crystals, a 0.003" chromel-constantan thermocouple was also attached to one of the faces with a non-electrically conductive paint.

The heat transfer "cell" comprised a two-liter, three-neck resin flask placed in a thermostatically controlled bath. The three inlets to the flask provided for the crystal holder, a gas inlet, evacuation and pressure measurement system, and a magnetically-coupled stirrer. The cell and its auxiliaries, then, permitted maintenance and measurement of the crystal voltage, control and measurement of the ambient temperature, variation of the gas composition, and measurement of the pressure. For those crystals provided with a thermocouple, the crystal surface temperature  $T_S$  was also available.

### Instrumentation

Electrically, the TANDEL may be thought of as a non-linear capacitor. That is, when in the autostabilized state, the current response of the device when subjected to sinusoidal excitation is distributed over several harmonics of the fundamental frequency. As the element moves from one autostable state to another in response to changes in any of the relevant variables already discussed, its impedance to current flow changes appreciably. This behavior suggests a bridge technique that will both maintain the autostable condition as well as permit monitoring these impedance changes.

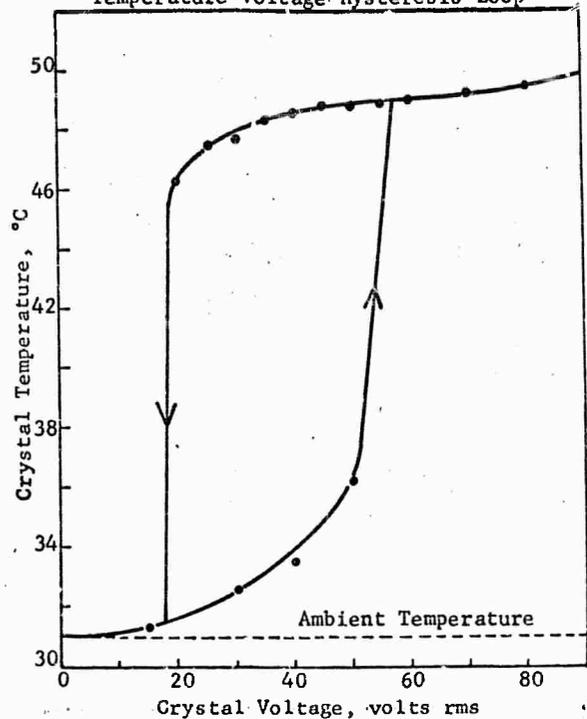
The experiments were performed by connecting the crystal into one arm of a General Radio, model 716-c, Schering-type capacitance bridge. A standard signal generator and a low-distortion, 10-watt amplifier provided the source of sine-wave power. Inasmuch as the bridge off-balance signal contained the same distribution of harmonics as mentioned above, a satisfactory null could only be achieved by filtering out all frequencies other than the fundamental. A Hewlett-Packard, model 310A wave analyzer was used for this, and an oscilloscope was used as a null detector. A panel meter on the wave analyzer also gave an indication of the off-balance signal and was used to obtain quantitative measurements during unbalanced operation of the bridge.

Crystal voltages were measured with a sine-reading rms VTVM, and crystal and ambient temperatures were determined from potentiometer readings of the respective thermocouple outputs. Temperatures reported are felt to be accurate to within  $\pm 0.1^\circ\text{C}$ . Variations in the cell temperature  $T_a$  due to cycling of the thermostat were observed to be less than  $\pm 0.05^\circ\text{C}$ . Measurements of the gas pressure in the cell were made with a mercury manometer.

### Confirmation of the Autostabilized State

Figure 1 presents data obtained on a TGS crystal fitted with a thermocouple for measuring its surface temperature. The temperature-voltage hysteresis phenomenon is apparent, and the region of high-temperature stability lies in the vicinity of the Curie point ( $49.4^\circ\text{C}$ ). The critical voltages for transitions to and from the autostabilized state varied widely from sample to sample. This, however, was not unexpected since these transition points clearly depend upon extensive rather than intensive properties of the crystal, and no attempt was made in this work to insure precise crystallographic uniformity among the TGS samples used. Nevertheless, the existence of the region of high-temperature stability in the vicinity of the Curie point was reproducible among all probes studied, and this is taken as confirmation of the autostabilization effect.

Figure 1  
Temperature-Voltage Hysteresis Loop



In the low-temperature state the crystal behaves similarly to an ordinary capacitor of 15 to 25  $\mu\text{F}$  with a dissipation factor on the order of  $10^{-3}$ . When the transition to the autostabilized state occurs, these properties increase dramatically, and the equivalent parallel resistance of the element decreases so as to maintain the heat balance at the new steady state. Now, as the crystal moves from one steady state to another in response to changes in the thermal ambient, both this resistance  $R_{xp}$  and the equivalent parallel capacitance  $C_{xp}$  vary from point to point. One can follow this process, then, by noting variations in either of these quantities from the successive bridge readings at balance. Furthermore, the heat generation term  $q$  at each steady state may be determined as the ratio of the square of the rms heating voltage to the parallel resistance.

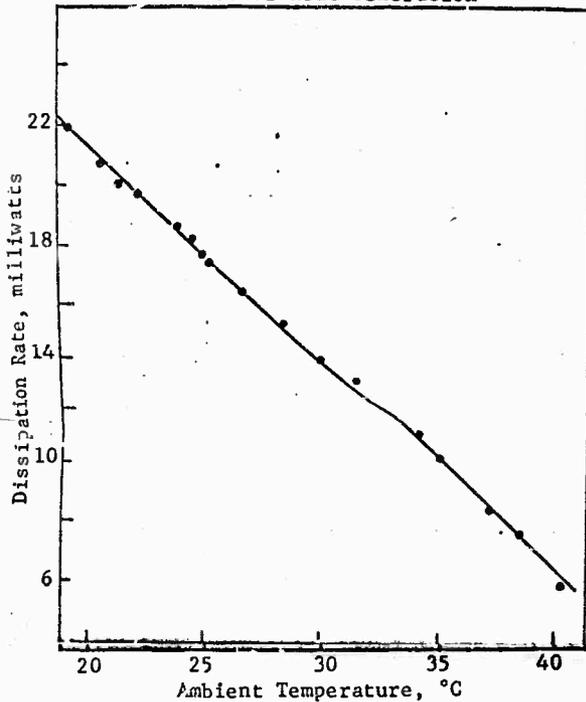
### Applications to Heat-Transfer Studies

Within the framework of the discussion presented earlier, the utility of TGS TANDEL as a heat-transfer gauge may be demonstrated by noting the extent to which its performance is consistent with equation (1). The data required for such a comparison were obtained by bringing the element into the autostabilized condition, changing the independent variable of interest (either  $T_a$  or some variable other than  $T_a$  that influences the heat-transfer coefficient  $h$ ), rebalancing the bridge under conditions of constant heating voltage, and noting the bridge readings.

### 1. Sensitivity to Changes in $T_a$

Data obtained in such a manner as to reflect the influence of variations in  $T_a$  are shown in Figure 2 for experiments in an atmosphere of  $CO_2$  at a pressure of 800 mm Hg and with an applied 20 kHz heating voltage of 70 V. The plot shows the dependence of heat generation upon ambient temperature over a range of  $T_a$  in excess of 20°C. Since the crystal used for these measurements had been fitted with a thermocouple for surface temperature determination, the values of  $T_s$  corresponding to each of the plotted points were also available.

Figure 2  
Rates of Heat Generation



The linearity of the data is in excellent agreement with the stability of TGS TANDEL predicted in the previous section. A more quantitative measure of the autostabilized crystal's ability to maintain its surface temperature relatively constant is obtained by noting that  $T_s$  varied monotonically during these measurements between 47.0 and 48.2°C. This suggests an overall stability for these measurements of about 0.06.

The range of  $T_a$  investigated here was considerably less than the total range over which one might expect the condition of autostabilization to prevail. This was simply the result of limitations of the particular instruments used in this set of experiments. In the previously cited work by Dvořák et al, stability data are presented for bulk temperature differences in

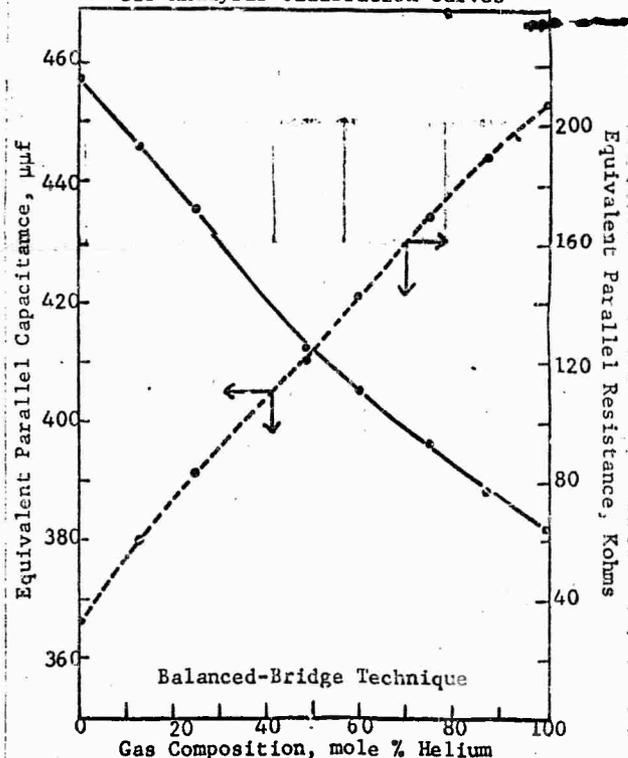
excess of 70°C at this same amplitude and frequency of heating voltage. Even under these extreme conditions where one might expect increased free convection to influence the linearity of the data, these authors report no significant variation in  $S$ .

### 2. Sensitivity to Changes in $h$

Due to the fundamentally conceptual nature of the heat-transfer coefficient, this quantity cannot be controlled directly (in contrast to  $T_a$ ), but rather, indirectly through its dependence upon the thermal conductivity and state of convection of the medium. It is, therefore, probably not meaningful to perform the analogous stability analysis for the  $h$ -varying case as was done previously. But in view of the likely future applications of the TANDEL as a transducer for  $h$ -influencing variables, it was felt appropriate to present data for one such typical case here.

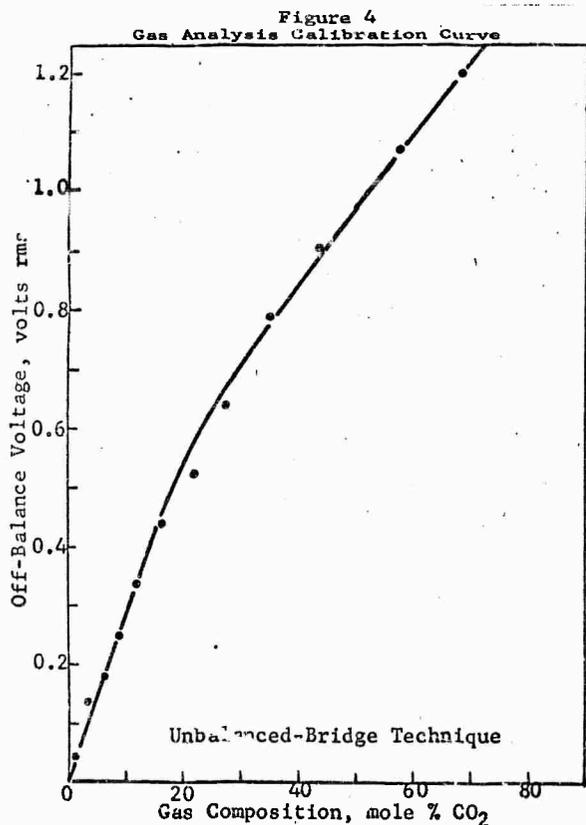
Figure 3 shows the results of an experiment wherein the composition of the gas was varied throughout the entire concentration span between pure carbon dioxide and pure helium. All other variables were held constant. At each concentration studied the bridge was balanced, and the values of  $C_{xp}$  and  $R_{xp}$  were determined.

Figure 3  
Gas Analysis Calibration Curves



These results reflect the variation in heat exchange between the crystal and its surroundings as the effective thermal conductivity of the gas changes from mixture to mixture. If the TANDEL were viewed as a transducer for concentration in binary gas mixtures, either of the functions plotted in Figure 3 might serve as a calibration curve (as might, of course, the computed value of  $q$ ). The sensitivity of the method, however, is uniquely determined by the minimum change in concentration that produces a detectable change in the bridge null signal. Although this was found to vary somewhat with concentration, and more dramatically among the several crystals studied, a conservative estimate of the sensitivity with the present instrumentation would place the minimum detection limit at 0.1 mole percent.

An alternative to the above method, and one which might find application in the detection of small amounts of a foreign material in a carrier gas, follows when the bridge is balanced with pure carrier in the cell and then permitted to move away from balance as the concentration of the contaminant builds up. This situation was simulated by considering pure helium to be the "carrier" and carbon dioxide the "contaminant." Figure 4 shows the results obtained at a frequency of 20 kHz, a constant heating voltage of 70 V and an ambient temperature of 31.1°C. The ordinate plotted is the amplitude of the fundamental component of the bridge off-balance signal as indicated by the panel meter on the wave analyzer.



The linearity of the data for CO<sub>2</sub> concentrations below about 17 mole percent is probably fortuitous. The slope of this line, however, in conjunction with the wave analyzer's detection threshold of about 0.5 mv, leads one to assign a minimum detection limit of about 0.015 mole percent. This sensitivity, although encouraging, is characteristic of the specific gas-pair and concentration range studied here. One may not conclude from these data that the unbalanced-bridge technique offers inherently greater sensitivity than the method discussed previously.

### 3. Accuracy and Sensitivity

The absolute accuracy of TGS TANDEL in measurements such as those reported here is meaningful only in reference to determinations of  $q$ . The other applications suggested (as well as some suggested by other authors) depend upon calibration techniques wherein a more appropriate statement of merit relates to sensitivity and reproducibility. However, it is clear that as a transducer for heat transfer, the accuracy is limited only by the instrumentation used for the measurement of electrical dissipation. In the present study, heat generation was determined from measurements of the crystal voltage with a sine-reading rms VTVM and measurements of  $R_{xp}$  with the capacitance bridge. To the extent that the voltage signal across the crystal contained higher harmonics of the fundamental excitation frequency, these determinations could have been slightly in error. Although the ultimate in dissipation accuracy was not felt to be important in this work, instrumentation exists to obtain  $q$  values at whatever level of accuracy is required.

A more important question concerns the magnitude of the measured heat transfer rates in comparison to those predicted for bodies of similar geometry. In an attempt to make this comparison, dissipation rates for a body of approximately equivalent geometry were predicted for conditions similar to those under which one of the TGS probes operated. The calculated and measured rates agreed to within about 25 percent. This level of agreement was felt to be quite satisfactory in view of the rather imprecise model upon which the calculations were based. The TANDEL was assumed in these calculations to be represented by an equivalent sphere losing heat to the surroundings through purely conductive and free-convective processes. No correction was introduced for heat loss through either the lead wires or the T<sub>g</sub> thermocouple.

The previous statements regarding the sensitivity of the ferroelectric transducer when used as a gas analyzer must also be viewed in terms of the present instrumentation system. The Schering bridge used in this work was operated in the substitution mode with the generator and detector connections interchanged. This method of operation was necessary to account for stray impedances not associated with the TANDEL and to maintain the excitation level

needed for the autostabilized state. The variety of experimental conditions investigated with the TGS probes in this study pushed the General Radio 716-c to its safe design limit with the result that certain data in the high-voltage, high-frequency region could not be reported. It is apparent that a more optimal bridge design would extend the useful range of this device and, perhaps, lead to a greater effective sensitivity.

It should also be kept in mind that single-ended operation need not be the only mode for this device. One might, as is the case with purely resistive probes, set up a differential arrangement utilizing two TANDEL probes, one exposed to a reference ambient and the other in contact with the system under study. The common-mode rejection associated with all such similar differential transducers would certainly result in greater sensitivity. The null-detection techniques for the bridge would, no doubt, be more involved in this case because of the complicated interaction of harmonics from the two crystals, but the basic idea seems sound.

#### Transient Response

No quantitative data are known to be available on the transient response of TGS TANDEL. A prediction of this might be made if one knew the temperature distribution both inside the crystal and in the surrounding gas together with such parameters as the relative thermal diffusivities of the two media. Before such an analysis is attempted, however, it is felt that a more thorough understanding of the actual physical processes taking place in the crystal is needed. Properties such as the specific heat vary in a very complicated manner in the vicinity of a second order phase transition, and it is not immediately obvious that the characterization of TANDEL as it moves from one steady state to the next can be accomplished by conventional methods.

Nevertheless, the TGS crystals used in this study exhibited no "apparent" lag in their response to "sudden" changes in the ambient conditions, so it was felt that a qualitative experiment was in order. With an active TANDEL in the cell at steady-state conditions and the bridge balanced, the magnetically-coupled stirrer was turned on, thus increasing the effective value of  $h$  through the added forced convection. The bridge was observed to move rapidly out of balance with the off-balance signal tending to stabilize at some new value. This rather crude method of inducing time-dependent behavior did, in fact, yield fluctuations simply because of the inconstant stirrer speed. This unsteadiness was reflected by variations in the off-balance signal from the bridge about the average value.

A second "pseudo-balanced" experiment was performed wherein the stirrer was set at a given speed, the bridge balanced at some mean condition and the excursions of the off-balance signal about the null point observed. Here again,

the response appeared to be fairly rapid, and insofar as the average speed of the stirrer remained constant, the pseudo-balance null did not vary. When the stirrer speed was changed, either downward or upward, the bridge moved rapidly away from this pseudo-balance, again tending to stabilize at some mean value of off-balance output. Insofar as the several stirring speeds were reproducible during the experiment, the corresponding time-averaged off-balance signals were similarly invariant.

A more quantitative evaluation of the transient response of TGS TANDEL will, of course, require more sophisticated experimentation. The probe must either be exposed to known disturbances or its performance must be compared with a device having known characteristics such as a hot wire. There will certainly be preferred physical configurations for a device used as a transducer for transient phenomena as opposed to the simple forms taken by the crystals in this study. It is still felt, however, that the response observed in these simple experiments is encouraging and justifies additional research.

#### CONCLUDING REMARKS

The applications of the ferroelectric transducer described in this paper, particularly because of the temperature auto stabilization effect, are far reaching in the fields of heat transfer and fluid dynamics. The marked increase in temperature stability that these devices offer promises to yield sensors for the linear detection of diffusion-controlled processes in gaseous systems, both flowing and at rest. It is interesting to note that an analogous technique has been developed for detecting diffusion-controlled processes in liquid systems wherein convected mass transfer is taking place. Known as the diffusion-controlled electrode technique and developed by Hanratty and co-workers (8,11,12), this experimental method has yielded significant information on the structure of a variety of flow fields. Much of the experience gained in the development of this latter technique will be of value as possible future applications are planned for TANDEL.

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