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VIBRATIONAL ENERGY TRANSFER IN EXPANDING MIXTURES OF N_2 AND CO_2 AS MEASURED BY AN ELECTRON BEAM

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16. Abstract

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SUMMARY

The transfer of molecular vibrational energy was studied by means of the electron-beam technique in expanding mixtures of N_2 and CO_2 heated by an electric arc. High-temperature flowing N_2 was mixed in a plenum chamber with CO_2 at total pressures of 5 to 10 atm (507 to 1013 kN/m^2) and total temperatures of 1500 to 4000 K and then expanded supersonically in a conical nozzle to free-stream conditions of about 0.05 mm Hg (7 N/m^2) and 150 K. Vibrational temperatures of N_2 were measured in the expanded flow and compared with temperatures predicted by using a time-dependent technique for the numerical solution of the nonequilibrium flow. By means of this technique and the experimental data, the vibrational relaxation rates of expanding N_2 and CO_2 were deduced. The results indicate generally faster rates than those measured in shock tubes for the vibrational-translational exchange in N_2 and for the vibrational-vibrational exchange between the asymmetric stretching mode ν_3 and the bending mode ν_2 of CO_2.

INTRODUCTION

The near-resonant transfer of vibrational energy from N_2 to the asymmetric stretching mode ν_3 of CO_2, illustrated in figure 1, has long been recognized as the process that makes N_2-CO_2 laser action possible (ref. 1). As a result of considerable research effort on the N_2-CO_2 molecular system (refs. 2 to 6) the general mechanisms involved are understood reasonably well, but the proper rate equations remain somewhat in doubt. The present objective is to measure directly the vibrational temperature of the N_2 molecules in a relaxing N_2-CO_2 mixture in order to evaluate these rates. Vibrational relaxation times are deduced from the measured N_2 vibrational temperatures by using a time-dependent technique for the numerical solution of the nonequilibrium flow of N_2-CO_2 mixtures through a convergent-divergent nozzle. In this technique the temperature dependence and magnitude of the relaxation rates can be varied until the N_2 vibrational temperatures predicted by the numerical solution agree with the measured temperatures. (See ref. 7.)

Since the gasdynamic laser using rapidly expanding N_2 and CO_2 has been shown to be practical (ref. 8), a knowledge of the mechanism of vibrational energy transfer within the modes of the various molecules and the rates that govern these transfers is necessary for prediction of laser performance. These rate processes are also important in nonequilibrium-flow calculations for vehicle entry into planetary atmospheres.

SYMBOLS

E	vibrational energy
H	enthalpy
M	collision particle
$N_2^+B^2\Sigma_u$	excited ion state of N_2^+
$N_2X^1\Sigma_g$	ground state of N_2
$N_2^+X^2\Sigma_g$	ground state of N_2^+
p	pressure
T	temperature
t	time
v	vibrational quantum number
x	distance downstream on center line of nozzle
X	mole fraction
ν_1	symmetric stretching mode
ν_2	bending mode
ν_3	asymmetric stretching mode
τ	vibrational relaxation time

τ_A,τ_B,τ_C	vibrational relaxation times illustrated in figure 1

Subscripts:

I,II	modes of N_2-CO_2 vibrational model
t	total
v	vibrational state

Superscripts:

e	equilibrium
*	excited state

EXPERIMENTAL METHOD

This investigation was carried out in the Langley 1-foot hypersonic arc tunnel, illustrated in figure 2. Flowing N_2 is heated as it passes through the magnetically rotated arc and it is then mixed with unheated CO_2 in the stagnation chamber. The mixture flows through a 0.55-cm-diameter sonic throat and expands in a 5^o half-angle conical nozzle to the test section. A vacuum system consisting of a five-stage steam ejector maintains a back pressure of 0.05 mm Hg ($7\ N/m^2$). The reservoir temperature is essentially constant for each test. Reservoir temperatures between 1500 and 4000 K were obtained for individual tests by changing the arc gap length with various outer electrodes and by varying the relative percentages of N_2 and CO_2. For a constant mass flow rate, stagnation reservoir pressure varied from 5 to 10 atm (507 to $1013\ kN/m^2$) for this temperature range. This pressure variation is included in the theoretical predictions. Reservoir enthalpy (and thereby temperature) was computed by the sonic-throat method. A calibration of this technique with a tube calorimeter which accepted the total gas stream flowing from the tunnel throat established the accuracy of the enthalpy determination within ±10 percent, as shown in figure 3. An explanation of the sonic-throat method and a detailed description of the tunnel and calorimeter are available in references 9 and 10. High-purity-grade commercial nitrogen (99.9-percent pure) and carbon dioxide (99.8-percent pure) were used, and a previous experiment (ref. 11) showed that similar impurity levels did not affect the measured vibrational temperatures in an expanding flow.

The limitations and errors of the electron-beam technique for measuring N_2 vibrational temperature have been discussed in references 12 to 15; therefore only the basic concepts will be described here. In this method an electron gun produces a narrow column of electrons (15 kV) which passes normal to the flow and through the test section (fig. 2): These fast electrons excite a small number of the particle making up the flow, and the emission of light resulting from the transitions of these excited particles to lower energy states is observed spectrographically to determine the N_2 vibrational temperature. For tests both with N_2 and with the CO_2-N_2 gas mixtures, the first negative system of N_2^+ dominates all other observed bands in the visible spectrum. The electron-beam excitation-emission path is given by

$$N_2X^1\Sigma_g \xrightarrow{\text{excitation}} N_2^+B^2\Sigma_u \xrightarrow{\text{emission}} N_2^+X^2\Sigma_g$$

and is illustrated in figure 1. Relative intensity ratios of the total emission from the (0,1) and (1,2) bands of N_2^+ determine the N_2 vibrational temperatures. References 13 and 15 present an expression for this intensity ratio as a function of T_V, and its applicability is a direct result of the Franck-Condon principle (ref. 16). This principle states that the electron transition in the molecule is so fast relative to the intermolecular motion that the internal motion of the molecule does not have time to adjust. Consequently, the electron-beam-excited N_2^+ spectra indicate the thermal distribution of the N_2 ground state. Lifetimes of N_2^+ emission bands have been measured and are in the range of 10^{-8} second. Quartz windows allowed observation of the light emission from the N_2^+ molecular ions along the beam path by means of a 1/2-meter spectrograph. Entrance and exit slits were adjusted so that the (0,1) and (1,2) bands of N_2^+ were just resolved in the first order by using a photoelectric readout which was fed to a micromicroammeter with output displayed on a strip recorder.

Some bands of CO_2^+ were observed, the strongest being located at 2883 and 2896 Å, but these bands were too weak for detailed resolution and therefore only the N_2 data were analyzed. Measuring the N_2 vibrational temperatures from relative band intensities by the electron-beam technique is synonymous with determining the relative populations of the N_2 vibrational levels. These measurements taken in a relaxing flow of N_2 can therefore be used to calculate the relaxation rate of N_2.

VIBRATIONAL RELAXING GASES

For every vibrational level of N_2 there is a corresponding near-resonant vibrational level of CO_2 in the asymmetric stretching mode ν_3. Therefore, in an expanding relaxing mixture of N_2 and CO_2, competitive processes will be occurring to depopulate the upper N_2 vibrational levels, and the observed relaxation time will be a net result of these exchanges. The vibrational-translational process between the N_2 molecules in pure relaxing N_2 was analyzed separately so that the competitive relaxing processes could be evaluated in the mixtures. A relative rational model of the many vibrational-translational and vibrational-vibrational energy exchanges which occur in a N_2 and CO_2 mixture has been presented by Anderson (ref. 7) and shown in figure 1. This diagram includes only the vibrational energy levels of interest in this investigation. The collisional transition probabilities computed (ref. 5) and measured (ref. 2) for this system indicate not only the very fast, resonant vibrational energy exchange between the $v = 1$ level of N_2 and the (001) level of CO_2, but also a fast exchange between the (100) and (020) levels of CO_2 due to Fermi resonance. Vibrational energy is also rapidly transferred among the lower excited levels of the degenerate mode ν_2 in CO_2 because of the nearly equal spacing of these levels. Since these fast transitions tend to couple various energy levels, Anderson (ref. 7) grouped the participating levels into two modes (modes I and II in fig. 1) which are assumed to be in equilibrium within themselves but not with each other.

In reference 7 a time-dependent technique is used for a detailed solution of this model, with the assumption that the vibrational energies of each mode relax as a simple harmonic oscillator. Therefore,

$$\frac{d(E_V)_I}{dt} = \frac{(E_V)_I^e - (E_V)_I}{\tau_I} \tag{1}$$

and a similar equation applies for $(E_V)_{II}$ with a relaxation time τ_{II}.

The relaxation times τ_I and τ_{II} are averages which characterize the net rate of energy transfer into and out of modes I and II and are governed by the transitions shown in figure 1 with relaxation times τ_A, τ_B, and τ_C. These relaxation times are also averages based on collisions between CO_2-CO_2, CO_2-N_2, and N_2-N_2 and are obtained from the "parallel resistance" mixture rule given in references 7 and 17:

$$\frac{1}{\tau_A} = \frac{X_{CO_2}}{\tau_A(CO_2\text{-}CO_2)} + \frac{X_{N_2}}{\tau_A(CO_2\text{-}N_2)} \tag{2}$$

Similar relations may be expressed for τ_B and τ_C. Two vibrational temperatures $(T_V)_I$ and $(T_V)_{II}$ are defined by using this model, where $(T_V)_{N_2} = (T_V)_{II}$. The distributions of the two vibrational temperatures along the nozzle center line are compared with the translational temperature distribution in figure 4 for conditions typical of this experiment. As shown, mode II, and therefore $(T_V)_{N_2}$, relaxes much slower than mode I, and neither mode I nor mode II is in equilibrium with the local translational temperature.

RESULTS AND DISCUSSION

Vibrational temperatures of N_2 were measured in expanded flows of pure N_2 and mixtures of N_2 and CO_2 as a function of total temperature. The results obtained in pure

N_2 are shown in figure 5, along with curves of the predicted values of T_V from the analytical program. Vibrational relaxation times used in computing the upper curve were taken from a line fitted to the shock-tube data for N_2 found in reference 2, and this distribution does not agree with the vibrational temperatures measured in the expanding N_2. To evaluate the data of this report, the relaxation-time expression is assumed to have the well-known form

$$\tau p = A \exp\left(BT^{-1/3}\right) \tag{3}$$

where p is the pressure and the constants A and B are evaluated by using the nonequilibrium program. The expression for τp that best fits the measured N_2 vibrational temperatures of figure 5 is

$$\tau p = 3.4 \times 10^{-12} \exp\left(195T^{-1/3}\right) \text{ atm-sec} \tag{4}$$

Vibrational relaxation times of N_2 observed in expanded flows are known to be less than predicted by the theory or measured in shock tubes (ref. 18), and several explanations for this difference are offered in references 19 to 21.

The analysis of the N_2-CO_2 mixture is somewhat more complicated in that six vibrational relaxation expressions must be considered. There are two expressions each for τ_A, τ_B, and τ_C; one expression is for collisions with like molecules and the other for collisions with unlike molecules. (See fig. 1.) The relaxation time $\tau_B(N_2\text{-}N_2)$ evaluated for expanding N_2 and given by equation (4) is used for analyzing the mixture and because of a lack of reliable data showing any significant difference (ref. 2), $\tau_B(N_2\text{-}CO_2)$ is set equal to $\tau_B(N_2\text{-}N_2)$. The relaxation rate of the CO_2 transition $(000) \longrightarrow (010)$ has been measured, by a number of techniques, for collisions with both CO_2 and N_2, and the results indicate that CO_2 is 4 times as effective as N_2 in this energy transfer process. (See ref. 2.) However, no measurements have been made of these relaxation times in an expanding flow. Therefore the values predicted by the Schwartz, Slawsky, and Herzfeld (SSH) theory are used for $\tau_C(CO_2\text{-}CO_2)$ and, as indicated in reference 2, $\tau_C(CO_2\text{-}N_2)$ is assumed to be equal to $4\tau_C(CO_2\text{-}CO_2)$. The difference in these relaxation rates is not critical to the present measurements because both the measured and predicted times τ_C are so much less than either τ_A or τ_B that the vibrational temperatures of the N_2, and therefore the relaxation time τ_A, will not be affected.

The greatest uncertainty lies in the selection of the values of τ_A, and the evaluation of these times is the primary purpose of this experiment. Taylor and Bitterman (ref. 2) assume that the ν_3 and ν_2 modes of CO_2 are coupled through the mechanism described by

$$CO_2^{*}(\nu_3) + M \rightleftarrows CO_2^{***}(\nu_2) + M + 416 \text{ cm}^{-1} \tag{5}$$

and most of the data presented by them indicate a rate from 10 to 100 times faster than that predicted by the SSH theory (ref. 5). Furthermore, the efficiency of CO_2 as a collision partner in this reaction is seen to be 2 to 3 times that of N_2.

The vibrational temperatures of N_2 measured in three mixtures of N_2 and CO_2 are shown in figures 6, 7, and 8, along with curves of the values of T_v predicted by using Anderson's model of relaxation in an expanding flow (ref. 7). The upper curve in figure 6 was computed by using the rates given in reference 2, which are based largely on shock-tube data, and this curve does not agree with the measurements in the expanding flow. The distributions of vibrational relaxation time which predicted the measured values of $(T_v)_{N_2}$ are shown in figures 6, 7, and 8, and these predictions are seen to be consistent for gas mixtures of 10.8 to 30 percent CO_2 in N_2. To obtain the proper predictions, the τ_A values are varied from the SSH predictions until a distribution is found to fit the data. The relaxation times τ_A are found to be 1/100 as long as predicted by the theory. They are in general agreement with the data of reference 2, although somewhat shorter. The $\tau_A(CO_2\text{-}CO_2)$ times are set at 0.27 those of $\tau_A(CO_2\text{-}N_2)$, as indicated in reference 2. The SSH values referred to for τ_A are taken from reference 5, where Herzfeld calculated the rates for the collisional deactivation of mode ν_3 of CO_2 for a large number of possible vibrational-vibrational mechanisms. The rate used is the overall rate which is the sum of all these processes. Dissociation and recombination are not considered in the analysis of these measurements, although CO_2 dissociation would have some significance for the high-temperature data. A decrease in the CO_2 concentration should result in an increase in the effective relaxation time, and therefore an increase in the vibrational temperatures. The higher temperature data of figures 6, 7, and 8 appear to show this effect.

CONCLUDING REMARKS

The vibrational relaxation rates for expanding mixtures of N_2 and CO_2 were evaluated by using the electron-beam technique and a time-dependent method for the solution of nonequilibrium flow. The results show that the vibrational relaxation rate between the asymmetric stretching mode ν_3 and the bending mode ν_2 of CO_2 is 100 times as fast as predicted by the Schwartz, Slawsky, and Herzfeld (SSH) theory. The specific processes that cause the discrepancy in these rates are not known, but the very fast relaxation times indicate near-resonant reactions. Since the SSH prediction is an overall evaluation of all the known transitions between the ν_3 and ν_2 modes, the theory appears inadequate to explain the complex mechanism for intramolecular vibrational-vibrational energy transfer in polyatomic molecules.

Langley Research Center,
National Aeronautics and Space Administration,
Hampton, Va., August 20, 1971.

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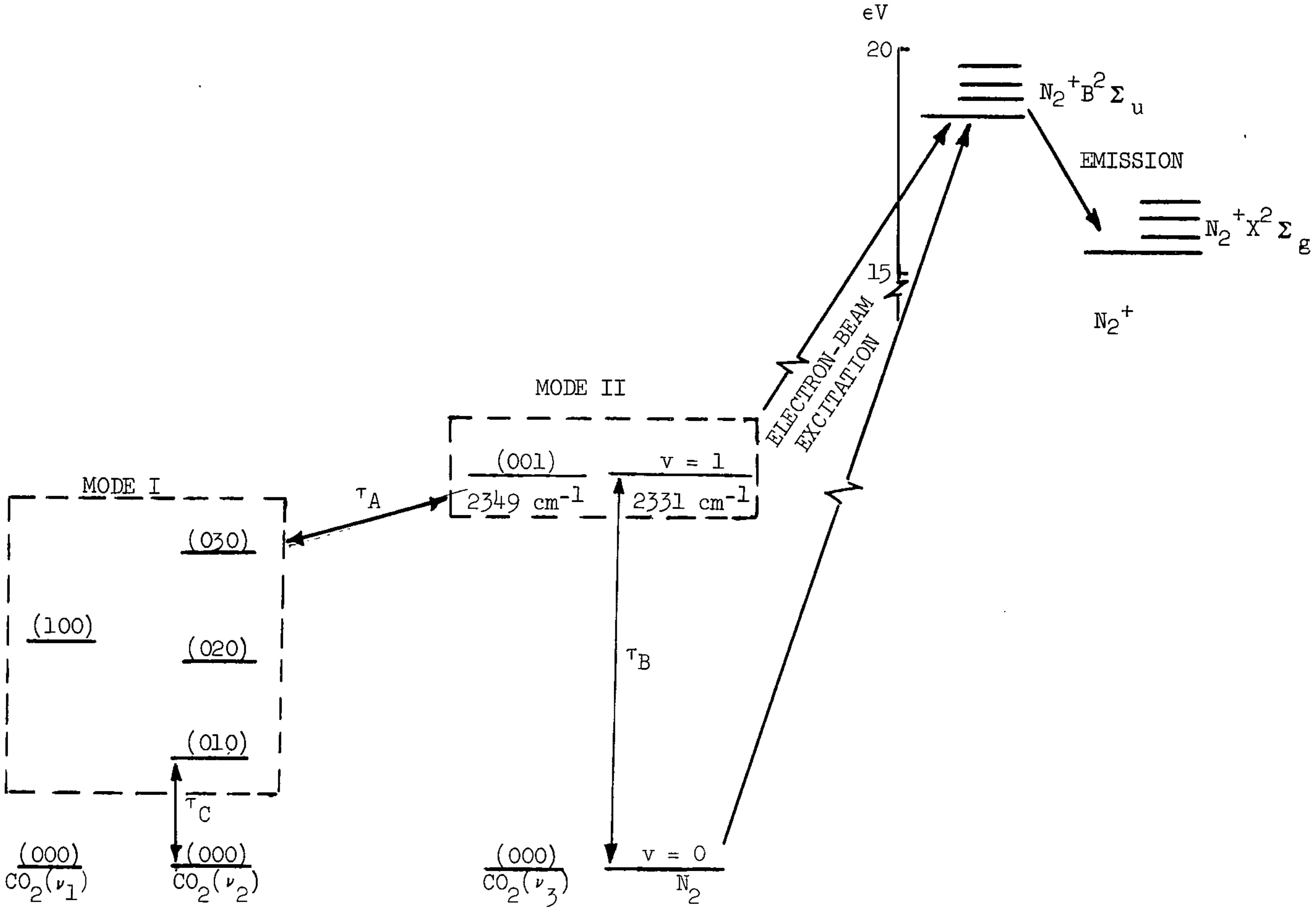

Figure 1.- Partial energy-level diagram for CO_2, N_2, and $N_2{}^+$ showing transitions important to the experiment.

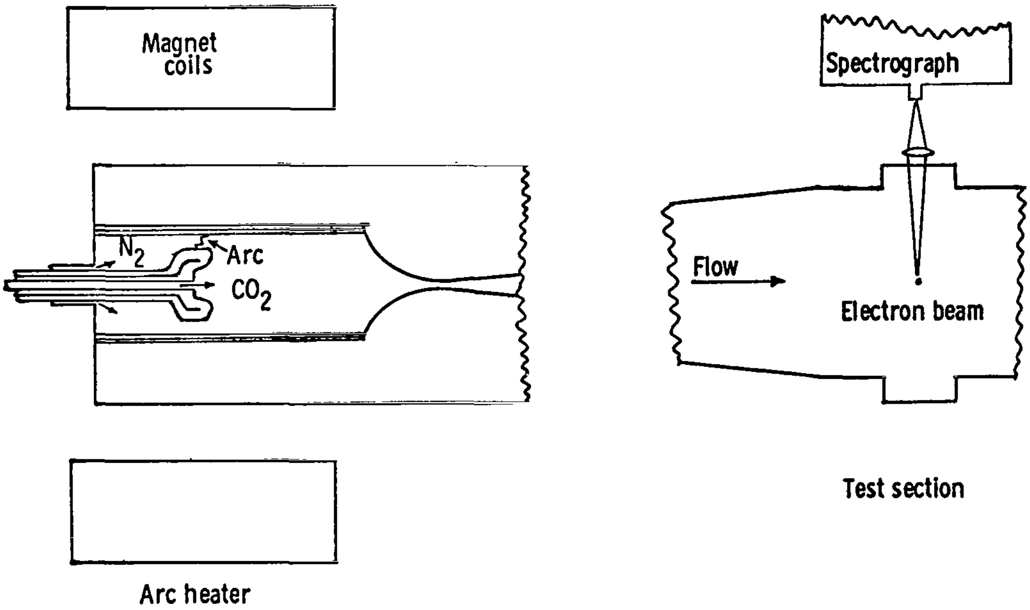

Figure 2.- Sketch of experimental apparatus and method used to inject gases into arc heater.

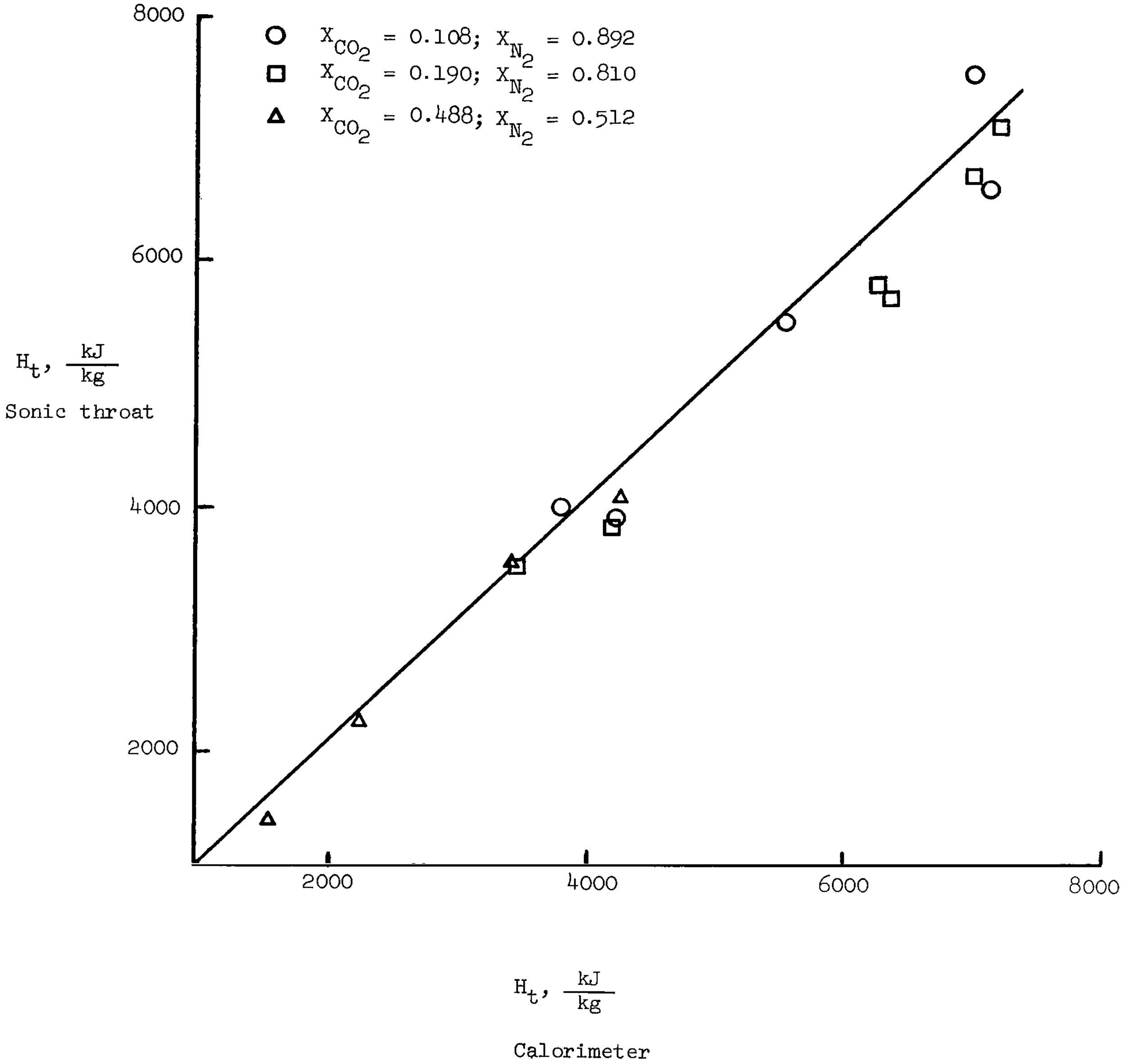

Figure 3.- Correlation of total enthalpy as determined by the sonic-throat method and by means of a calorimeter.

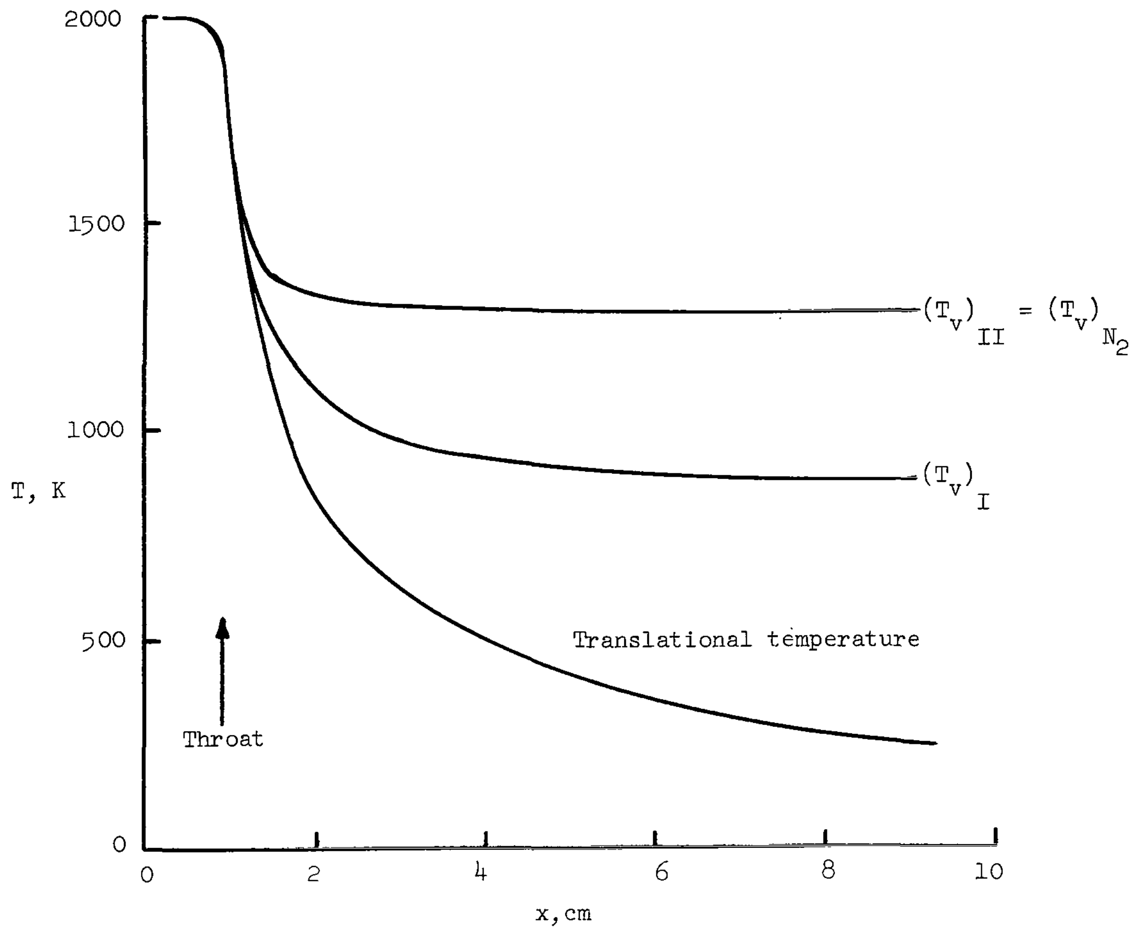

Figure 4.- Vibrational and translational temperature distributions through the nozzle for a mixture of $X_{CO_2} = 0.108$ and $X_{N_2} = 0.891$. $T_t = 2000$ K; $p_t = 5.8$ atm.

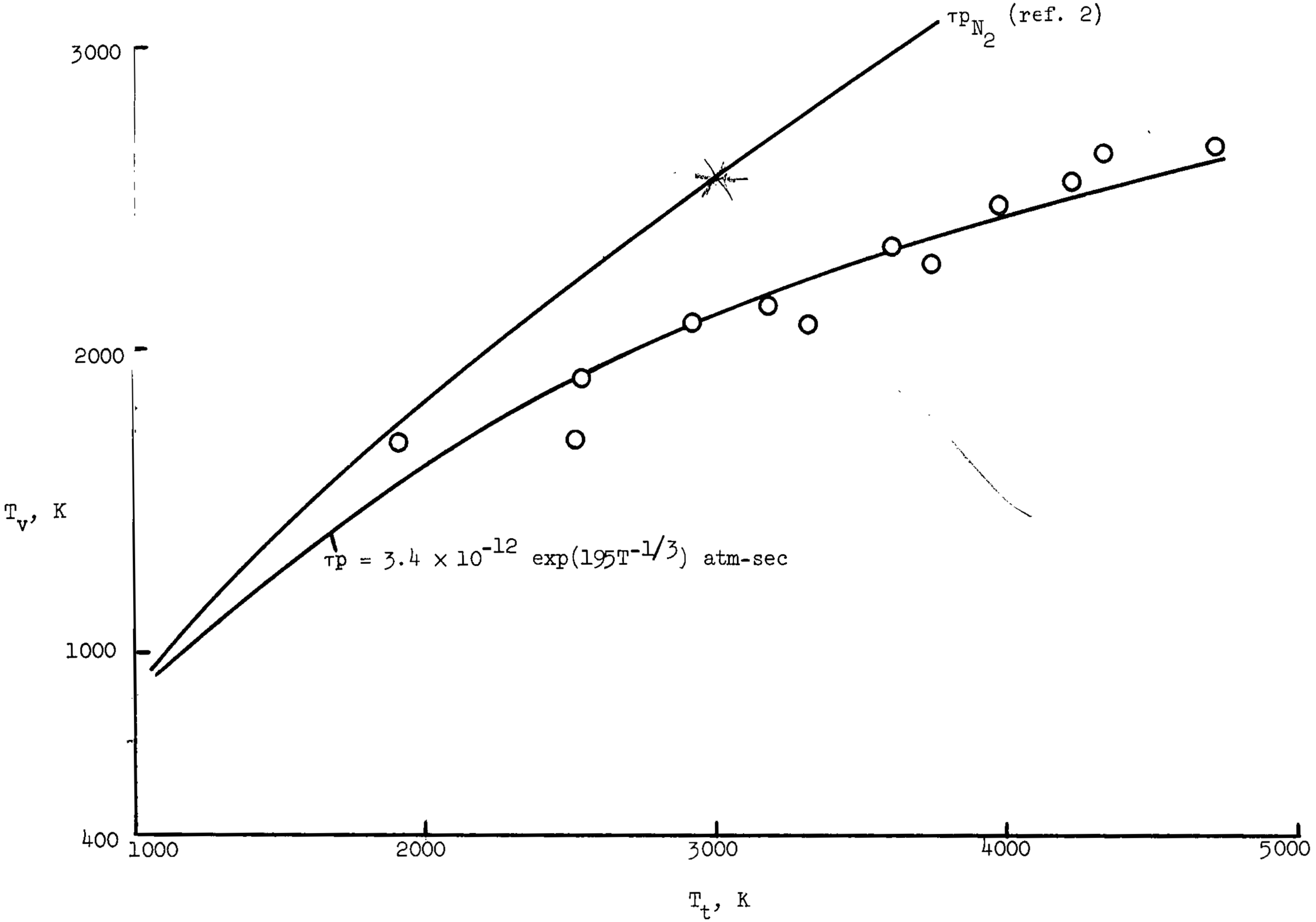

Figure 5.- Measured and predicted N_2 vibrational temperatures as a function of total temperature in pure N_2.

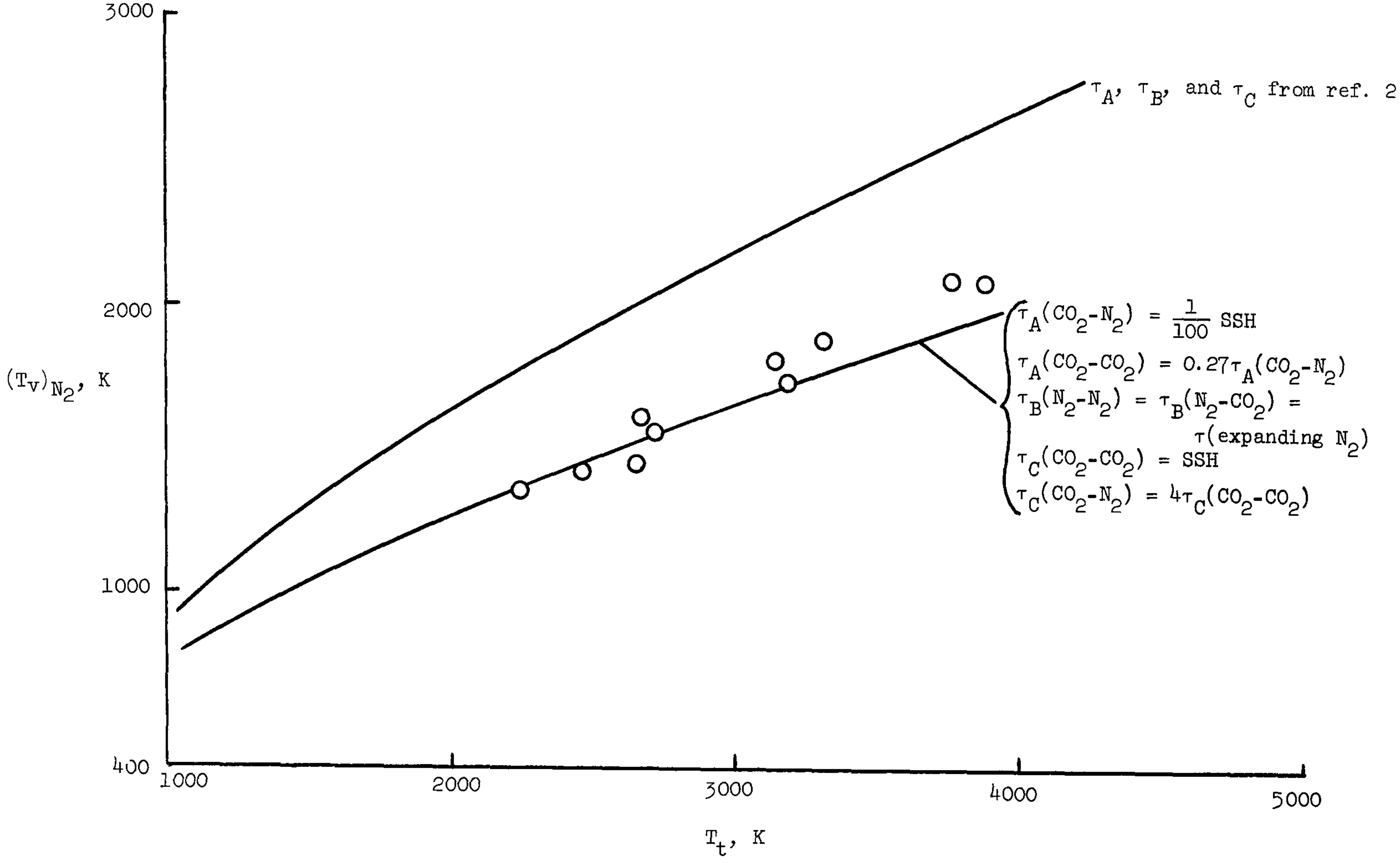

Figure 6.- Measured and predicted N_2 vibrational temperatures as a function of stagnation temperature in a mixture of $X_{CO_2} = 0.108$ and $X_{N_2} = 0.892$.

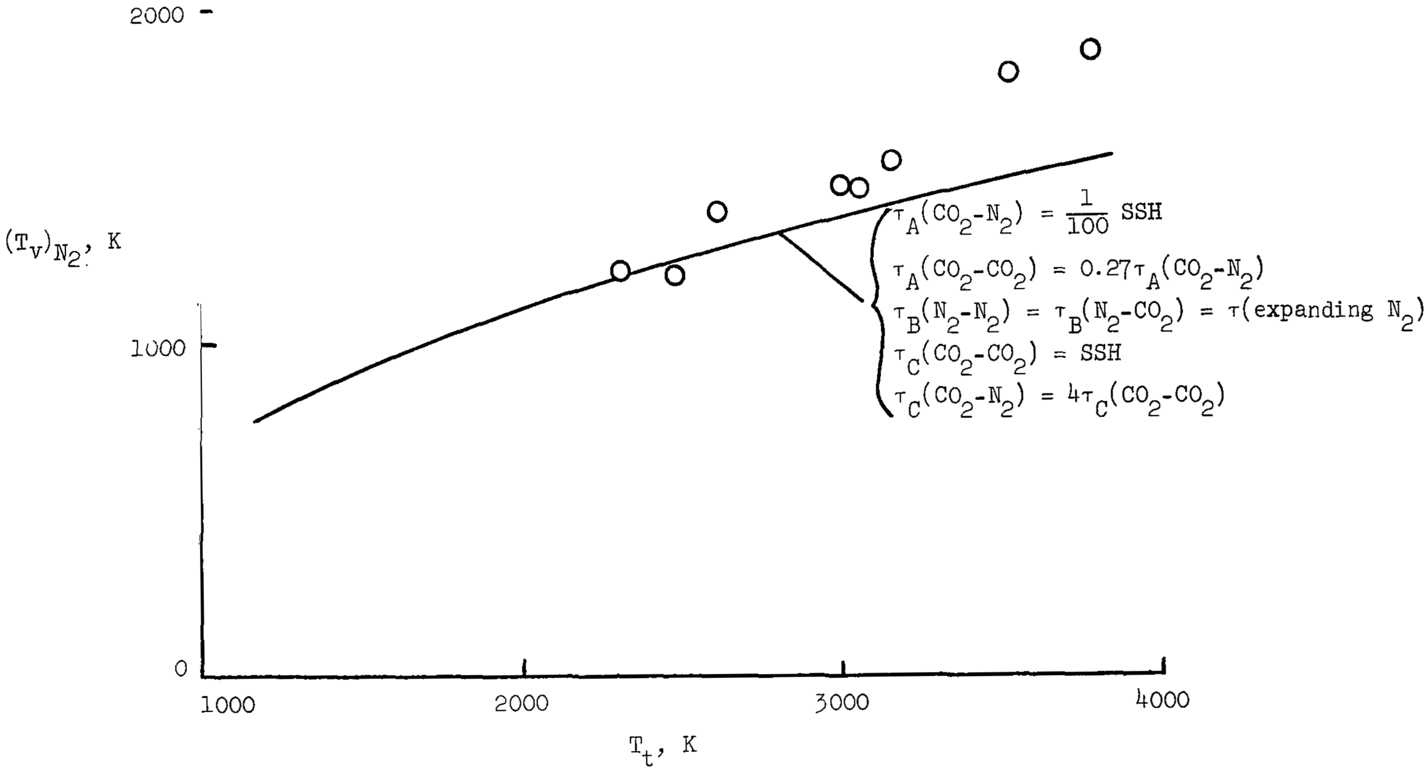

Figure 7.- Measured and predicted N_2 vibrational temperatures in a mixture of $X_{CO_2} = 0.190$ and $X_{N_2} = 0.810$.

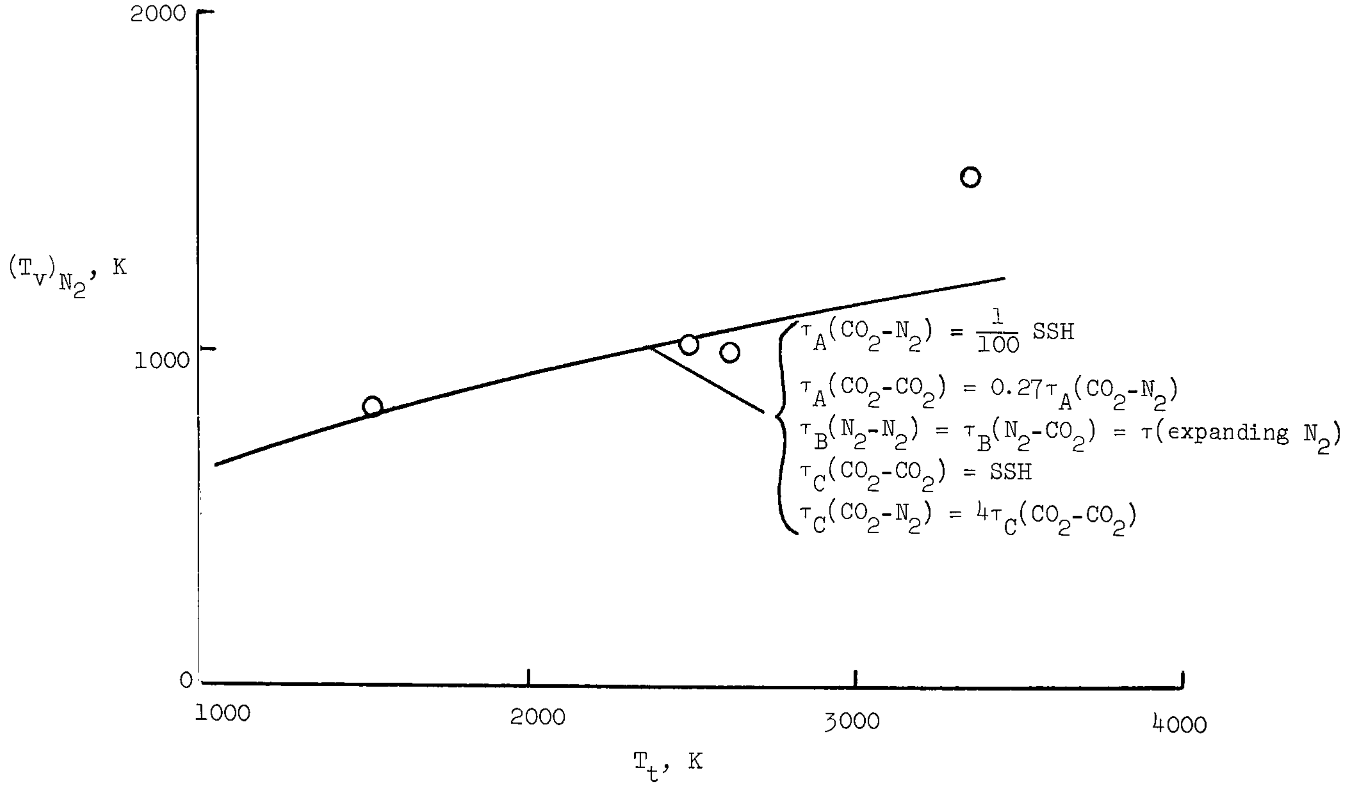

Figure 8.- Measured and predicted N_2 vibrational temperatures in a mixture of $X_{CO_2} = 0.300$ and $X_{N_2} = 0.700$.

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