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SPECTRUM-LINE-REVERSAL TEMPERATURE MEASUREMENTS THROUGH UNSTEADY RAREFACTION WAVES IN VIBRATIONALLY-RELAXING OXYGEN

by

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MINISTRY OF AVIATION, LONDON, W.C.2

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SUMMARY

Pressure and spectrum-line-reversal temperature measurements have been made of the structure of an unsteady expansion wave initiated in a vibrationally-excited gas in a shock tube by the rupture of a second diaphragm.

With oxygen initially at about 2500°K and 0·75 atmospheres pressure ($M_s \sim 6{\cdot}9$, $p_1 \sim 10$ torr), the vibrational temperature was observed to fall from its initial (fully-frozen) value at the tail of the wave to a minimum before rising again to the original equilibrium value at the head of the wave.

This agrees broadly with Appleton's calculations[11] based on a linear rate law and Landau-Teller theory for τ_v, although the temperature minimum was slightly higher and occurred later than he predicted; it has also been predicted qualitatively by Stulov[17].

More detailed studies with, for example, nitrogen would be desirable for seeking a full explanation of the small differences with theory revealed by these preliminary tests.

CONTENTS

ILLUSTRATIONS

1 INTRODUCTION

As part of a programme to investigate non-equilibrium and chemical kinetic phenomena in the flow of high-temperature gases, some shock-tube studies are being made of the structure of unsteady expansion waves in vibrationally-relaxing gases.

The purpose of this interim Note is to summarize the results of some preliminary tests done in oxygen; it is hoped to report more detailed measurements in nitrogen and in mixtures with other gases (for instance argon) at a later date.

Whenever rapid changes of translational energy occur in molecular gases (for example from passage through strong shock waves, combustion and detonation waves, or expansion waves), adjustment of their internal vibrational energy tends to lag behind on account of the relatively large number of collisions needed to excite or de-excite this mode. In high-speed flows, the particle residence time may be insufficient to allow a complete energy re-distribution between the vibrational and translational degrees of freedom and a 'non-equilibrium' flow results.

Experimental studies of vibrational relaxation effects in steady expansive flows have been made by several investigators. For example, Feldman[1] and more recently Cleaver[2] have employed wedges to generate Prandtl-Meyer expansions in air and in CO_2 respectively. Spectrum-line-reversal temperature measurements in expanded nozzle flows have been reported by Stollery[3], Stollery & Townsend[4], and recently by Hurle, Russo & Hall[5], in which direct evidence of vibrational energy 'freezing' in air and in nitrogen was obtained. The work of Hurle et al is particularly interesting in that it suggests that de-activation of vibrational energy states may proceed much faster than inferred from shock-wave activation studies. Bray[6] has recently reviewed theoretical and experimental studies of non-equilibrium phenomena in hypersonic nozzle flows including vibrational relaxation. Numerical computations of hypersonic nozzle flows with allowance for vibrational relaxation have been given by Stollery & Smith[7], and by Stollery & Park[8].

It is believed that the present work may be the first recorded attempt to employ an unsteady expansive flow to investigate a non-equilibrium phenomenon, in this case the vibrational relaxation of slightly-dissociated oxygen. In principle a much higher degree of expansion is possible than in the corresponding steady Prandtl-Meyer configuration, thus allowing tests over a wider temperature range.

The experiments were done in the same shock tube as used for earlier work[9,10], now modified by insertion of a second fragile diaphragm across the low-pressure section to form a constant area duct downstream. The unsteady flow pattern resulting from rupture of the second diaphragm by the shock-heated test gas is illustrated in the idealized x-t diagram of Fig.1. A backward-facing rarefaction wave, centred at the diaphragm station, propagates into the relaxing gas flowing into the expansion section; the temperature variation through this wave is measured at a fixed point using the spectrum-line-reversal method - there is much evidence for believing this indicates vibrational temperature

directly[5,21]. A simultaneous measurement of static pressure serves to check the flow geometry since it is insensitive to relaxation effects. Oxygen was chosen for the test gas in the first phase of the work since its vibrational relaxation time was short enough to allow observation of the expansion wave structure within the shock-tube running time.

In physical terms, the first particles to be observed behind the second contact surface have undergone a very rapid expansion to a low density and to low translational temperature and consequently there are insufficient collisions to transfer the excess internal vibrational energy they carry to the translational mode, i.e., the vibrational temperature is 'frozen' at the equilibrium value reached behind the primary shock. The subsequent increase in local density and translational temperature then causes a fall in vibrational temperature due to the rising collision rate and the increased time available for vibrational adjustment. Molecules observed in the region of the wave head will consequently be almost fully relaxed, and at the wave head itself the vibrational and translational temperatures will have finally become equal at equilibrium.

These experiments are complementary to some earlier numerical calculations by Appleton[11]. Fig.2 is a typical result based on his paper, showing the expected time variations of vibrational and translational temperature, and of static pressure measured at a fixed point. The distinctive profile of vibrational temperature will be noted and this has been confirmed in the experimental studies described below.

Although for simplicity these studies involved frozen chemistry (i.e. dissociation) extension to reacting gases with a view to measuring the temperature dependence of recombination rates is feasible and might incidentally show whether coupling between the rates of vibrational and chemical relaxation[12] is significant.

In the next section experimental arrangements for determining the structure of the expansion wave in terms of its profiles of spectrum-line-reversal temperature and of pressure are outlined, with only brief reference to the actual measuring techniques as these have been fully described in some previous work[9,10]. In Section 3 the results of a typical run are compared with computed profiles based on a conventional model of the internal relaxation process with no chemical reactions taking place[11,13]. Finally, in Section 4 the significance of the broad agreement found between the experimental and theoretical profiles is discussed and some suggestions made regarding further experimental studies.

2 EXPERIMENTAL ARRANGEMENTS

2.1 The shock tube

A schematic diagram of the arrangement of the shock tube for the expansion wave experiments is shown in Fig.3.

The driver and low-pressure sections were of 2-inch internal diameter and of lengths 5 ft and 15 ft respectively, selected to maximize running

time and minimize the shock attenuation as far as possible. For protection against corrosion the tubes were Kanigen nickel plated.

The constant-area expansion section was in the form of a brass tube about 2 ft long and 2-inch bore and with its inner surface lapped smooth.

A dump chamber 8 ft 6 in. long by 1 ft 6 in. diameter was used to avoid reflected disturbances in the expansion section.

2.2 The second diaphragm

Ideally this should open instantaneously without causing reflection of the primary shock, as suggested by the sketch of Fig.1. However, opening times of between 30 and 60 microseconds were indicated for the 0·0076 millimetre aluminium diaphragms used in these experiments. Fig.4 shows the results of some simple tests of aluminium foil and 0·013 millimetre Melinex diaphragms which gave opening delays of about 30 μs. Wilson's[14] calculated value for the aluminium foil was about 44 microseconds.

2.3 Test conditions

In the present work, hydrogen at room temperature was used as the driver gas and, in conjunction with 24 SWG scribed copper diaphragms bursting at 220 psi ±5%, gave primary shock Mach numbers measured at the second diaphragm position of about 7·0 in oxygen for an initial pressure of 10 torr (mm Hg). In the absence of attenuation effects and diaphragm opening delays, this would provide an initial equilibrium state of the oxygen with $T_2 \sim 2500°K$, $p_2 \sim 600$ torr, a degree of dissociation $Z_2 \sim 1\%$, (assumed frozen during the experiment), and about 14% of the total static enthalpy residing in the vibrational mode (the maximum possible).

The shock tube proportions must be so chosen that the reflected head of the rarefaction wave in the driver gas will not overtake the primary contact surface before the 'end point' equilibrium temperature of region 2 is observed at the head of the second expansion wave. At the same time, it is necessary to choose initial conditions that will make the time scale of the relaxation process less than the available running time.

In these experiments the duration of the unexpanded flow at the second diaphragm position was about 160 microseconds, and this was usually just sufficient to allow complete observation of the expansion wave profile.

For ease of comparison between theory and experiment it was desirable to reproduce the initial conditions closely in successive runs. The high- and low-pressure sections were therefore evacuated before being filled with the driver and test gases respectively. Even so, the primary shock speeds were reproducible only to about ±3% probably due partly to diaphragm opening and to attenuation, particularly when ignition had occurred at the hydrogen/oxygen interface.

A further result of attenuation of the primary shock is that the initial conditions of the experiment cannot be defined by a single measured shock speed since particles entering the expansion wave at successively later times carry increasing enthalpies.

Consequently, in these tests the primary shock speed was monitored along the channel so that the effect of attenuation could be subsequently estimated by the method of Ref.10.

2.4 Temperature measurement

The temperature measurements were made by means of the double-beam spectrum-line-reversal method developed by Clouston, Gaydon and Hurle[15]. As the experimental details have been given previously[9,10], only brief remarks are made here.

In the present work, the Pointolite background temperatures were set at about 2000°K and 2400°K to bracket the expected variation of vibrational temperature. This resulted in an emission signal on one beam and an absorption signal on the other, and enabled the temperature variation through the expansion to be followed with better accuracy than by extrapolation from dual emission or dual absorption records.

As in the earlier work, the sodium D-lines were used and the same method for introduction of the sodium iodide smoke; reasonably controlled amounts (0·1% to 0·2% by particle number) and good dispersion of sodium in the shock-heated gas resulted.

The windows, of plane optical quartz $\frac{1}{4}$" in diameter, were flush-mounted and were cleaned thoroughly before each run. The time resolution was about 5 microseconds as before, being limited electronically to reduce the photo-multiplier noise to an acceptable level.

2.5 Pressure measurements

The static pressure variation was measured with a miniature lead zirconate piezo-electric transducer having a nominal sensitivity of 24 mV/(lb in^{-2}) and a rise time of about 10 microseconds - this was the 'top hat' gauge developed by Stevens and described in Ref.16.

Since the instant of opening of the second diaphragm could not be determined precisely, some form of alternative event marker was needed to indicate the 'start' of the pressure and temperature traces. On the latter traces the secondary shock precursor usually caused a small emission 'spike' which served this purpose; a corresponding pressure 'spike' however did not appear, possibly being masked by the rather long rise time of the transducer. However, a detectable signal was given by a flush-mounted thin-film resistance thermometer when the secondary shock passed, and in conjunction with the pressure signal on a dual-beam oscilloscope display enabled the 'start' of the pressure trace to be estimated.

2.6 Vacuum system

To obtain a 'full' expansion (i.e. to the low pressure behind the secondary shock), the working section and dump chamber were evacuated to between 10^{-5} and 10^{-6} torr by means of a 9 in. oil diffusion pump backed with a 900 litres per minute rotary pump.

The rest of the vacuum system, sketched in Fig.3, was the same as that used previously; connections to vacuum gauges, pumps etc., were positioned well away from the second diaphragm region.

3 RESULTS

About twelve runs in oxygen were recorded, the initial conditions being held as nearly as possible the same for all. Figs.5 and 6 show, respectively, typical results of a simultaneous measurement of line-reversal temperature and of static pressure in a single run.

Fig.5 shows an experimental oscillogram of the time variation of sodium D-line radiation intensity against background brightness temperatures of 2005°K and 2390°K on the upper and lower beams respectively, recorded at a distance of 15·5 in. from the second diaphragm station. Emission causes a downward deflection and absorption an upward deflection. The upper trace shows a gradually increasing emission signal commencing from the 'spike' due to the secondary shock marking the start or 'tail' of the expansion. The lower trace shows an absorption signal increasing to a maximum from this point and then falling. Both traces show a discontinuity with passage of the primary contact surface.

In the temperature analysis of Fig.5(b) a comparison is made between the experimental temperature history (open symbols) derived from Fig.5(a) and a theoretical vibrational temperature profile calculated by Appleton's method[11,13] for comparable initial conditions. For the purpose of this comparison, the time origin of the temperature oscillogram was taken to be the instant when the primary shock reached the second diaphragm, determined by extrapolation of the shock speed versus distance data.

There is a striking similarity between the profiles, suggesting that the observed spectrum-line-reversal temperature is essentially the same as the vibrational temperature of the oxygen molecules for the present experimental conditions. The experimental temperature minimum occurs, however, some 20-25 microseconds later than expected, and the temperature itself is about 250°K higher than the theoretical value; moreover, the time scale of the observed temperature variation is shorter than that of the theoretical. The significance of these small discrepancies is discussed later, but they may in part arise from the delay in rupture of the weak diaphragm, with a consequent modification of the flow geometry; this might be expected to affect the pressure variation as well.

Fig.6(a) is the corresponding oscillogram of the pressure and thin-film gauge signals at a point on the wall 12·5 in. from the second diaphragm. The pressure signal is plotted in Fig.6(b) (using the same time origin as previously) against the theoretical pressure variation[13] for very similar initial conditions. Here it is seen that the experimental pressure signal is also delayed, and its rate of rise greater than predicted and with a time scale again less than that calculated by Appleton.

4 DISCUSSION

The 'breakaway' and subsequent rise of the spectrum-line-reversal temperature towards a 'frozen' value observed in these experiments shows that whilst the electronic excitation temperature of the sodium is not coupled to the molecular translational temperature, its value is consistent with the supposition that it is the vibrational temperature of the relaxing oxygen that is being measured by this means.

The existence of a minimum value of the vibrational temperature beyond which it again increases, (first demonstrated theoretically by Stulov[17] for dissociational relaxation), is a characteristic feature of the wave structure, and therefore the present results lend at least qualitative support to the calculations of Appleton and Stulov.

A comparative simultaneous measurement of spectrum-line-reversal and vibrational temperatures under non-equilibrium conditions has not so far been attempted, but use might be made of heteronuclear molecules (e.g. CO or NO) in a shock tube and time-resolved absorption/emission spectroscopy.

On account of the low density, there was insufficient sodium radiation present in these experiments to permit accurate measurement of temperature as far back in the flow as the fully-frozen region, although the observed temperature began to climb towards it. Runs at higher initial pressure, which would have made this possible were, however, not attempted with oxygen because of the risk of ignition at the driving interface. Comparable runs in nitrogen at initial pressures of up to 100 torr should overcome this limitation since the D-line radiancy ($\propto p^2$) would then be greater by a factor of roughly $\sqrt{10}$. Together with the use of interference filters of increased transmission (factor of 3) and more sensitive photo-multipliers, now available, the overall signal-to-noise ratio could be improved by a factor of at least 10. This should not only enable detection of the 'frozen' vibrational temperature, but will incidentally extend the measurable temperature range to below 1800°K.

A further important experimental reason for raising the density is that the sodium radiation may be collision-limited in the 'frozen' region of the expansion leading in this case to spuriously high observed temperatures. This may arise if the excited vibrational levels of the oxygen molecules are depopulated by transfer of energy to the electronic levels of the sodium atoms which then radiate, rather than by the binary collisions assumed in the theoretical description. Here again, nitrogen is to be preferred to oxygen as a test gas since spectroscopic data for collisions between nitrogen and excited sodium exist[18].

Evidently Kirchoff's law will not apply in this region and an experimental indication of this was that meaningful gas temperatures could not be derived from the ratio of the double-beam signals. This small region, around the tail of the expansion wave, corresponds to the dashed portion of the experimental temperature profile in Fig.5(b).

Allowing for the diaphragm-opening delay already referred to, the general agreement between theoretical and experimental profiles obtained shows that the picture of the flow pattern derived from theory is broadly correct, though the degree to which the wave is centered has not been investigated; it is hoped to check this in later work in which measurements will be taken at two downstream positions simultaneously. To avoid spurious distortion of the temperature and pressure profiles due to shock attenuation use of a larger diameter shock tube would be desirable.

The observed shock attenuation would have led one to expect temperatures in the region of the contact surface above the equilibrium value, based on the shock speed at the second diaphragm position. In fact however, in all runs, values less than equilibrium were recorded (Fig.5(b)).

This may indicate that the running time was not quite sufficient for equilibrium to be reached, but may also be a further illustration of an effect peculiar to spectrum-line-reversal measurements employing sodium in hot oxygen observed in some earlier work and not yet explained[9,10,19]. It was found that whilst changes of temperature were accurately followed the temperature values were low by constant systematic amounts ranging up to nearly 400°K in some cases. This effect was not present with sodium in nitrogen.

Reference has been made earlier to the results of Hurle et al[5] wherein it was necessary to postulate a relaxation time for de-excitation of N_2 in a nozzle expansion fifteen times shorter than that derived from vibrational-excitation studies behind strong normal shock waves. From this they suggest that the Landau-Teller description of vibrational de-excitation may be inapplicable in non-equilibrium situations where the difference between vibrational and translational energy is large, and that the shorter relaxation time indicates that the probability of de-excitation may depend on the vibrational as well as on the translational energy of the colliding molecules.

The present results, however, suggest that for the oxygen relaxation observed in these experiments such assumptions are unnecessary. The general agreement seen with Appleton's calculations implies that his description of the relaxation process was adequate; he used a linear rate equation, Camac's experimental data (from shock-wave studies[20]), and Landau-Teller theory.

The consequences of large changes of the effective relaxation time on the temperature profiles of unsteady expansion waves in nitrogen have been looked into theoretically by Woodley[13] and some results are illustrated in Fig.7. A substantial increase in the rate of vibrational de-excitation (of the order of that mentioned by Hurle et al) will shift the temperature minimum to earlier times and reduce its value considerably, as might be qualitatively expected. This effect was not observed in the present experiments with oxygen, the results of which are consistent with a normal value for the effective relaxation time within a factor of about two. In fact, the experimental observation of vibrational temperatures above theoretical (Fig.5) might, on the contrary, suggest that de-excitation proceeds more slowly than was assumed. According to Appleton's argument[11], the assumption of frozen chemistry in these experiments is justified, so that the enthalpy of recombination of the oxygen atoms in the wave cannot contribute to the vibrational temperature. A possibility that was

considered was that these higher temperatures were due to reflected sodium light entering the photo-multipliers. However this seems unlikely since the aperture of the optical system was small, and earlier work[10] involving temperature measurements of the incident shock never at any time revealed a sodium radiation signal preceding the shock.

5 CONCLUSIONS

(1) Spectrum-line-reversal temperature measurements through an unsteady expansion wave in vibrationally-excited oxygen have demonstrated that the vibrational temperature falls from its initial fully-frozen value at the tail of the wave to a minimum, and then subsequently rises to the original equilibrium value at the head of the wave.

(2) This agrees broadly with Appleton's calculations and shows that a linear rate law and Landau-Teller theory can adequately describe the vibrational de-excitation even with large departures from equilibrium. The value of the effective relaxation time inferred from the experiments agree with accepted values within a factor of two.

(3) The small differences with theory revealed by these preliminary tests are thought to arise mainly from the time-delay in rupture of the second diaphragm. Temperature differences also arise from the use of Na in hot oxygen and from the observed shock attenuation. To reduce these effects, further tests will be made in nitrogen at higher initial pressures and with Cr instead of Na resonance radiation.

At the same time this will also avoid the possibility of collision-limiting of the radiation from the low-density, fully-frozen region of the expansion wave.

ACKNOWLEDGEMENTS

The author is indebted to Mr. J. G. Woodley for carrying out the numerical calculations of the temperature and pressure profiles shown in Figs.2 and 7.

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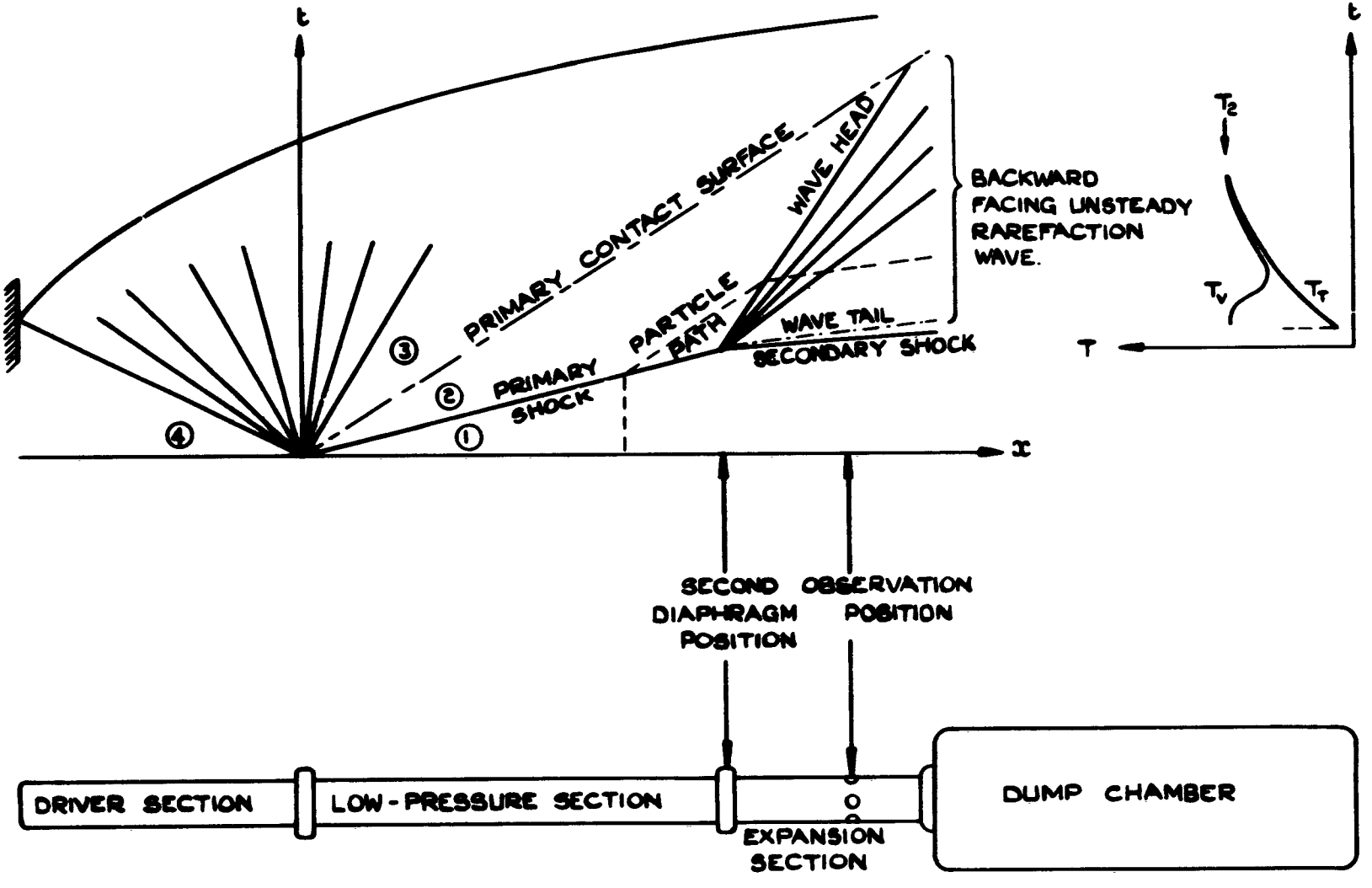


FIG. 1. SCHEMATIC DIAGRAM OF IDEALIZED FLOW GEOMETRY IN $x - t$ PLANE.

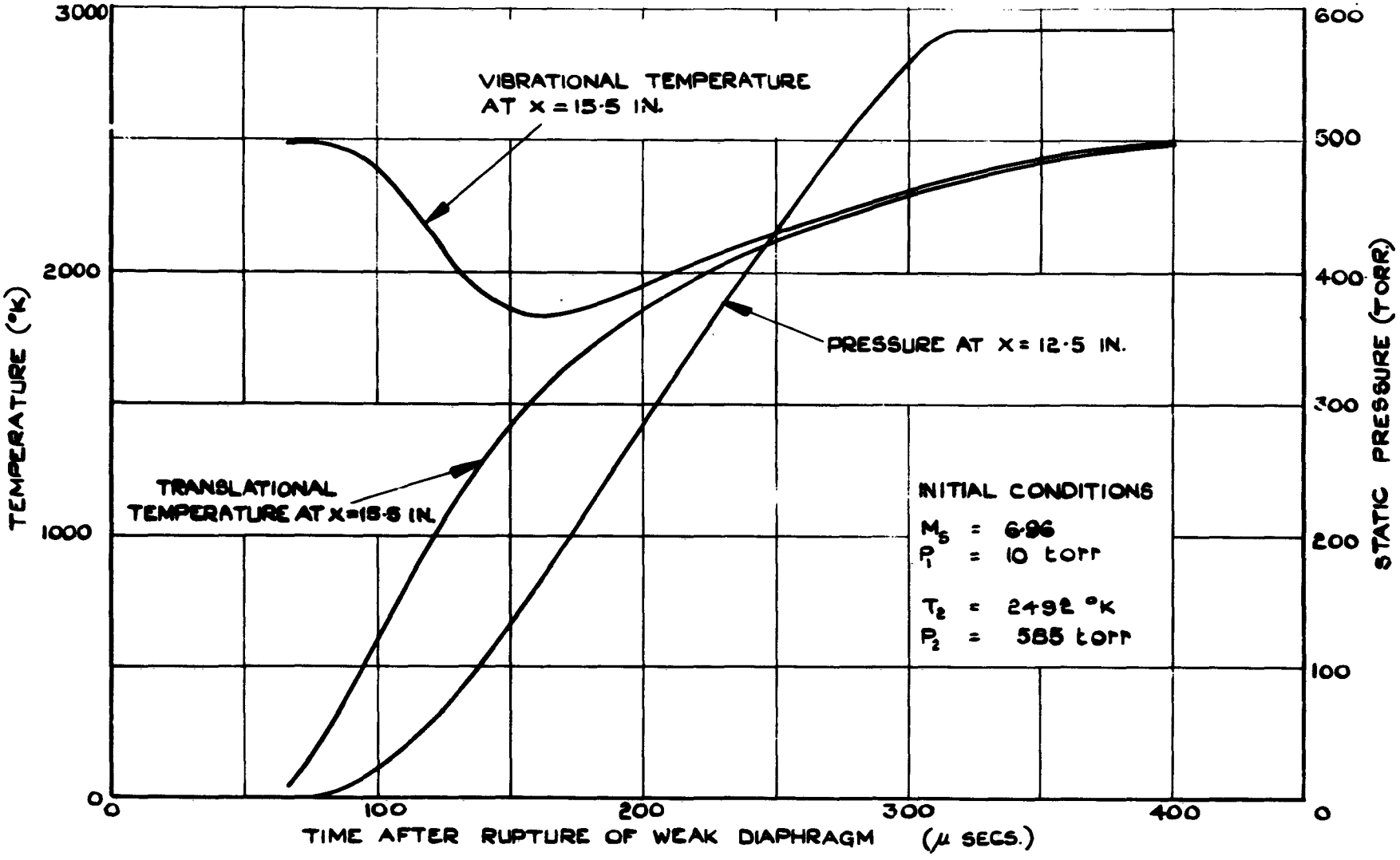


FIG. 2. CALCULATED TEMPERATURE AND PRESSURE VARIATION IN AN UNSTEADY EXPANSION WAVE IN PURE OXYGEN (AFTER APPLETON")

T.N. AERO. 2968. FIG. 2.

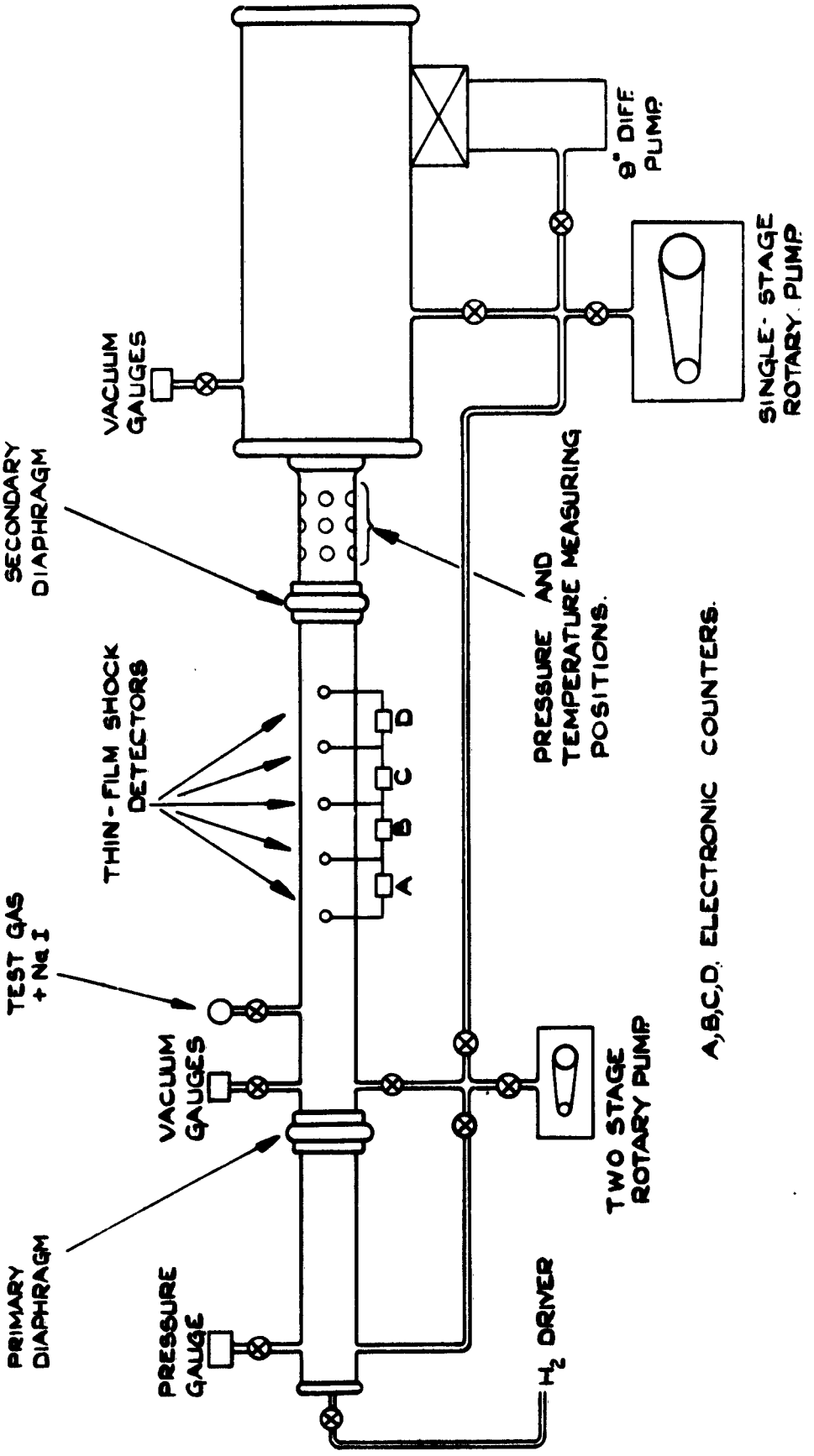


FIG. 3. DIAGRAMMATIC LAYOUT OF 2" DIAMETER SHOCK TUBE FOR EXPANSION WAVE EXPERIMENTS.

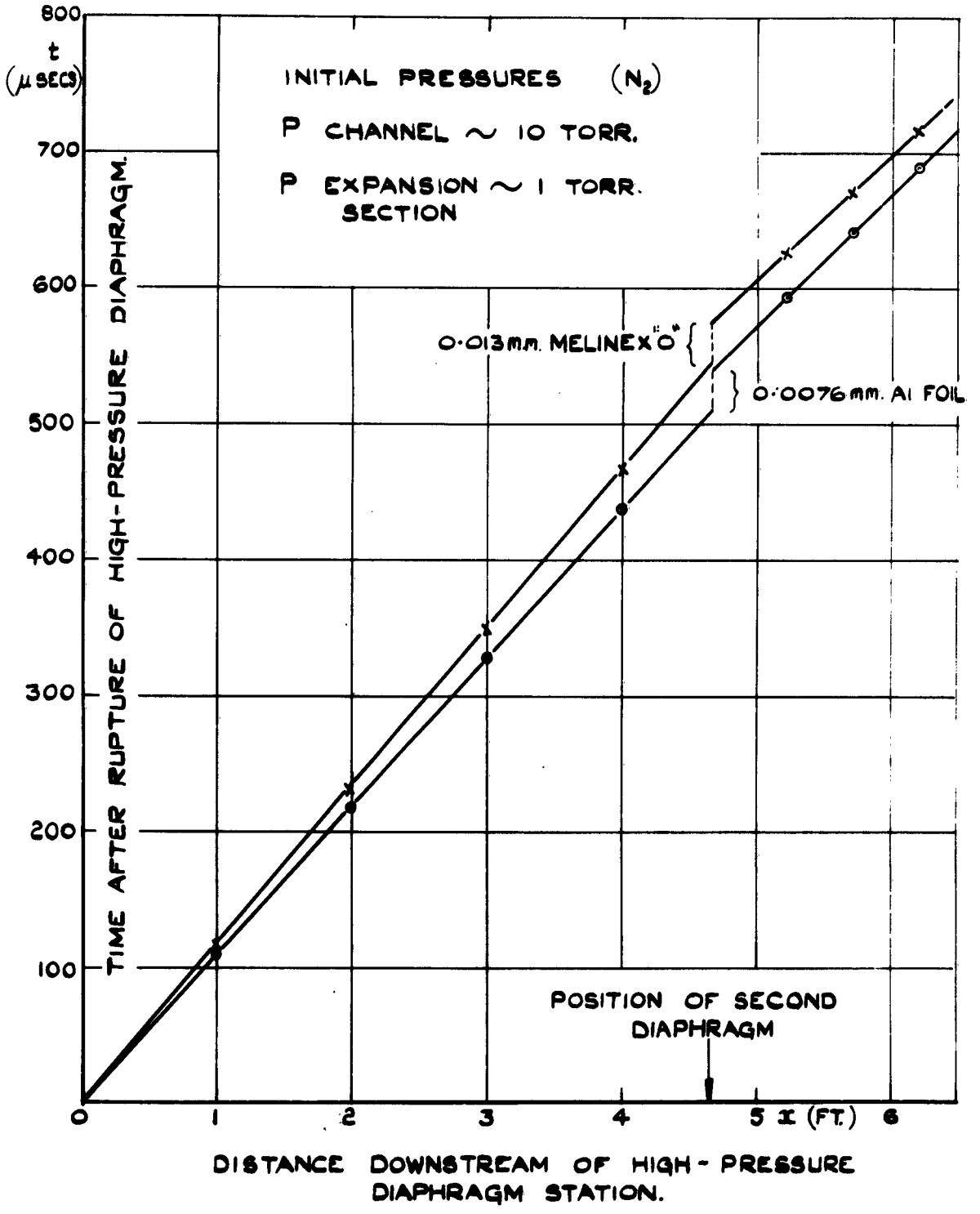
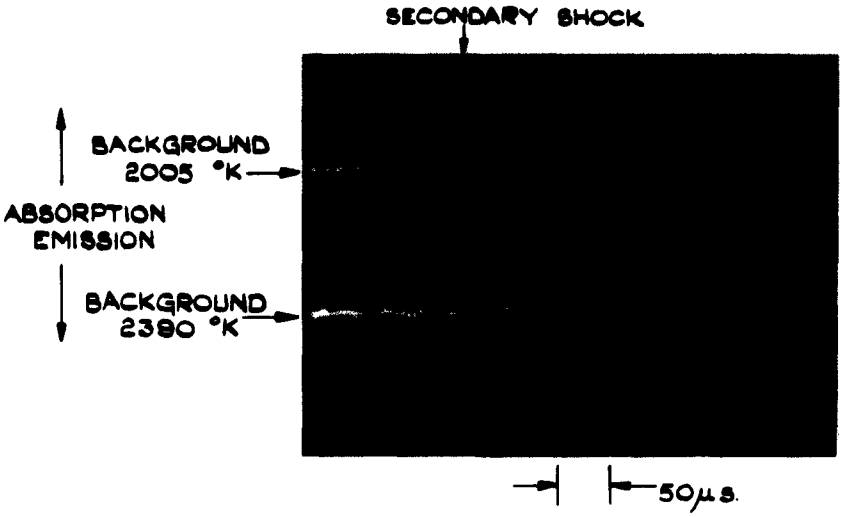
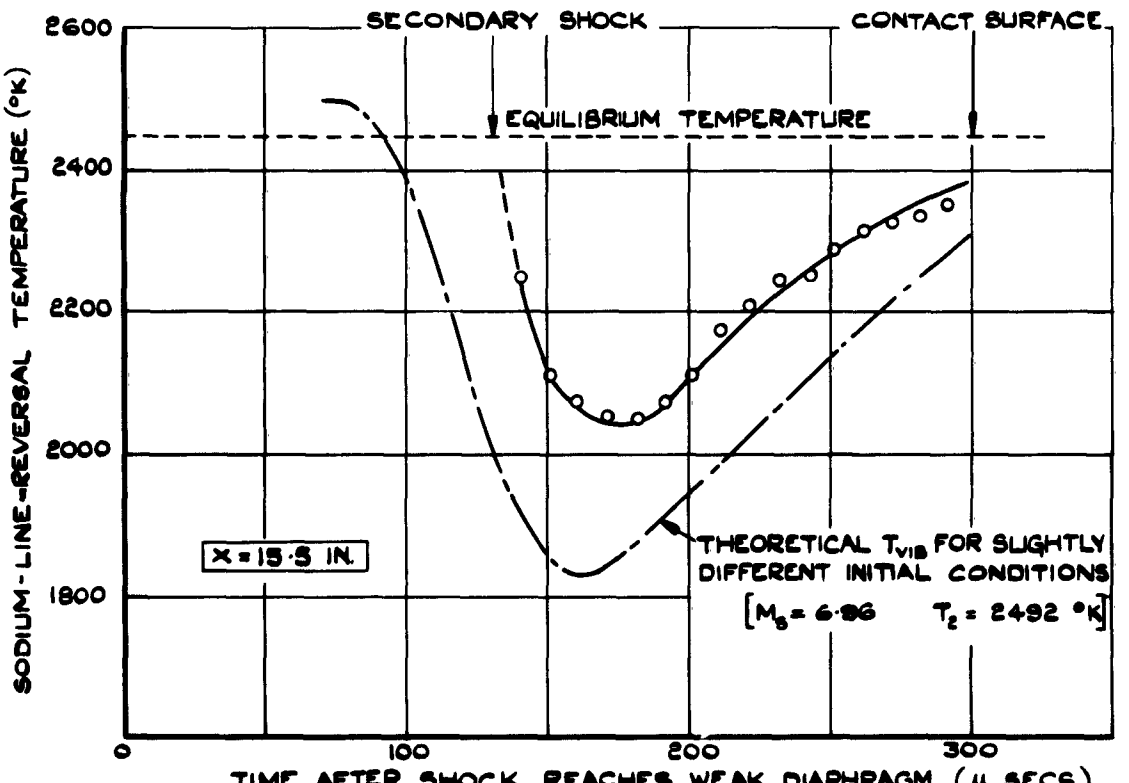


FIG. 4. DERIVATION OF SECOND DIAPHRAGM OPENING TIMES.

FIG. 5. (a & b)



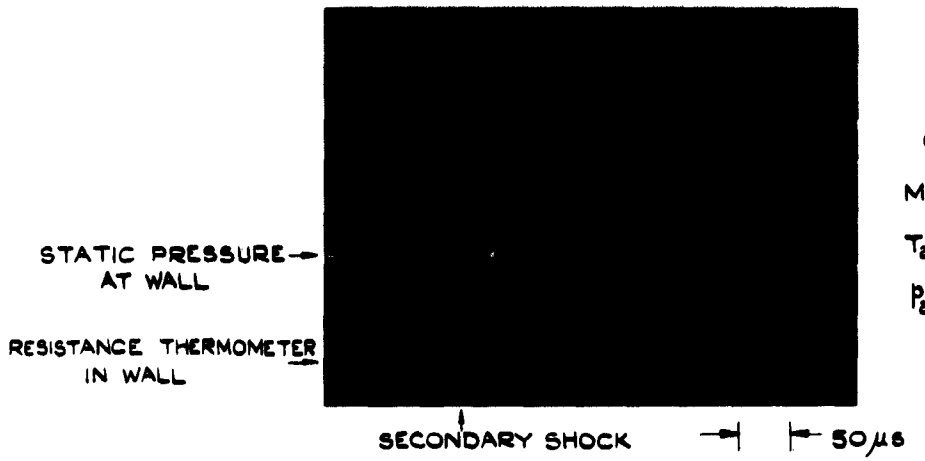
(a) TYPICAL DOUBLE-BEAM RECORD.



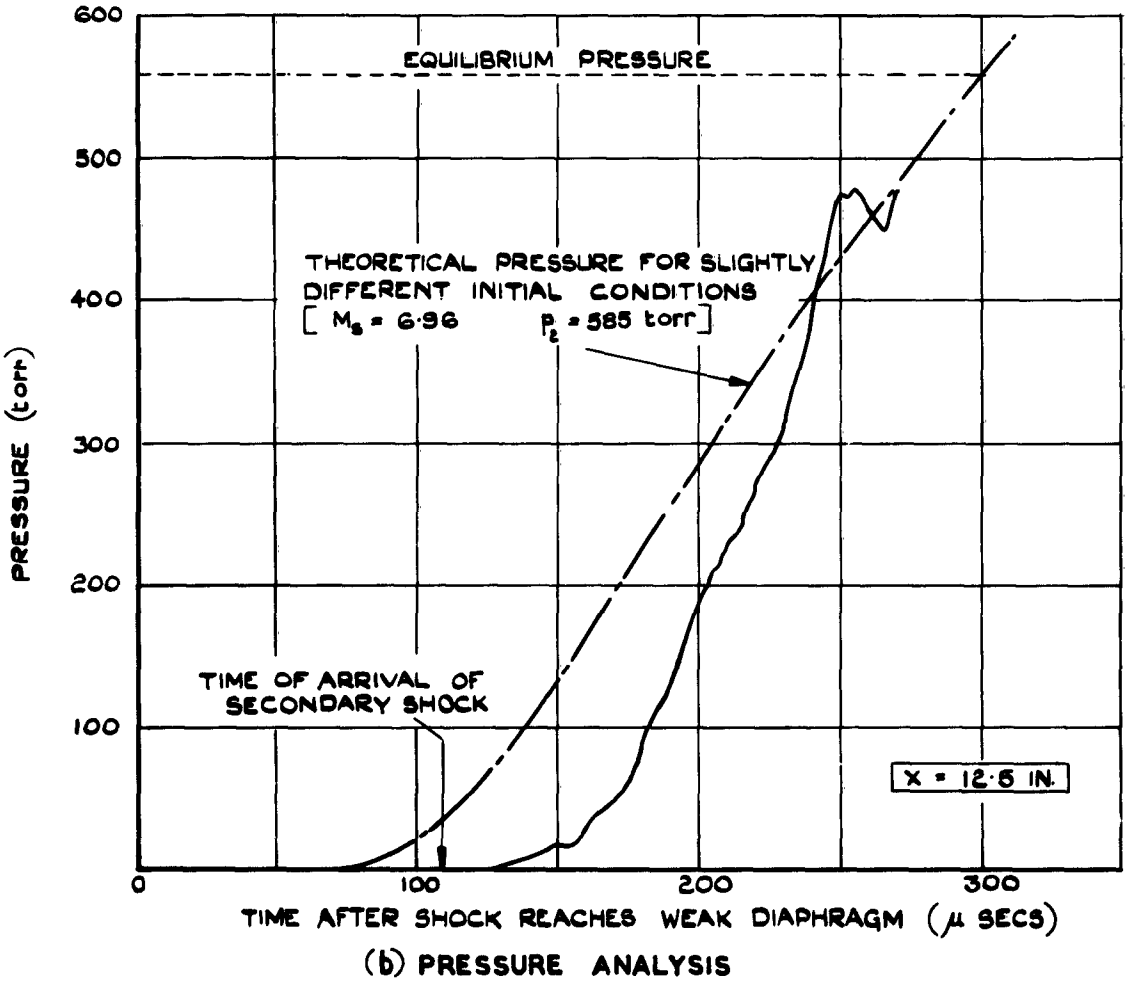
(b) TEMPERATURE ANALYSIS.

FIG. 5. (a & b) OBSERVED S.L.R. TEMPERATURE VARIATION THROUGH AN UNSTEADY EXPANSION WAVE IN SHOCK-HEATED OXYGEN.

FIG. 6. (a & b)



(a) TYPICAL OSCILLOGRAM.



(b) PRESSURE ANALYSIS

FIG. 6(a & b) OBSERVED PRESSURE VARIATION THROUGH AN UNSTEADY EXPANSION WAVE IN SHOCK - HEATED OXYGEN.

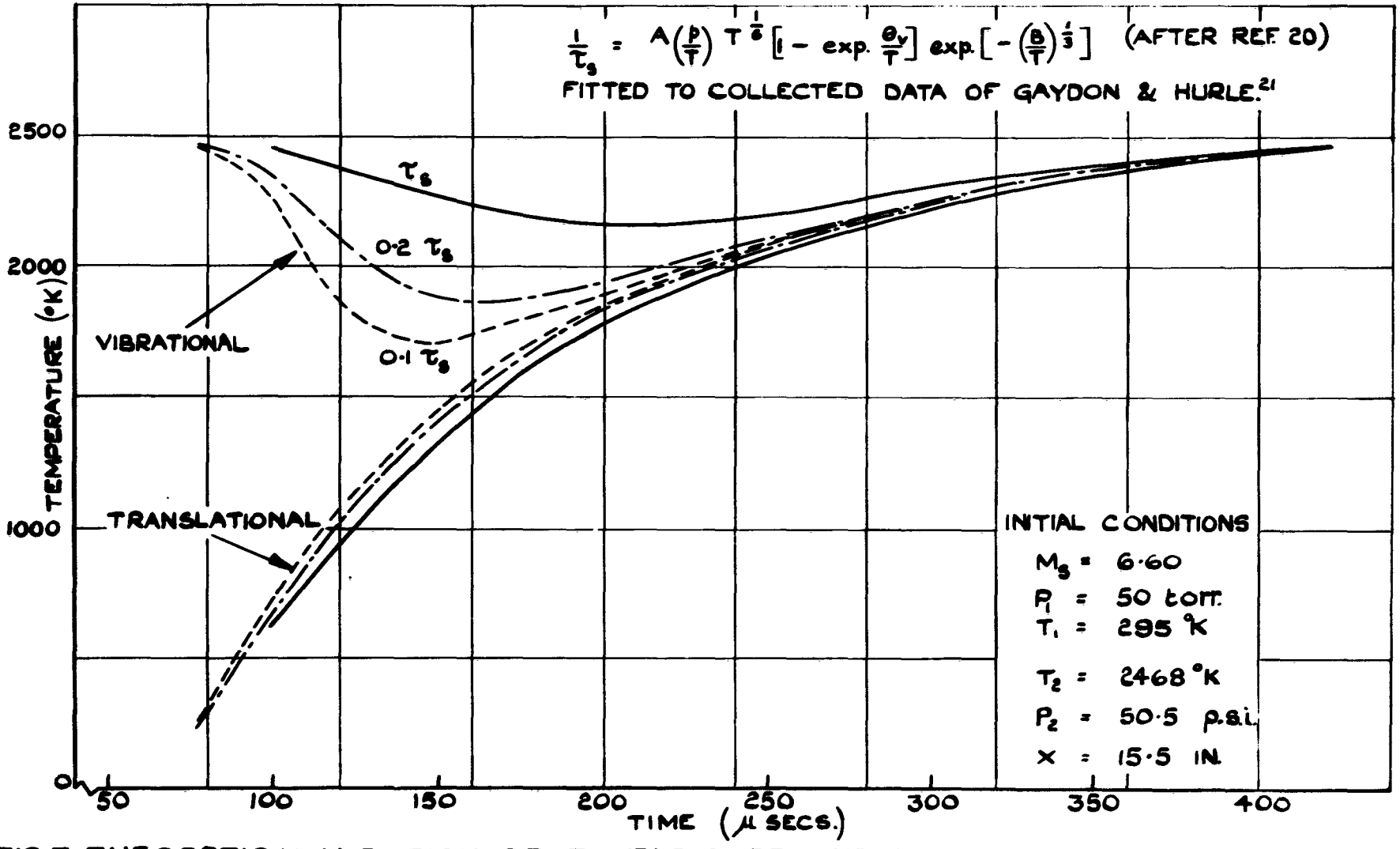


FIG. 7. THEORETICAL VARIATION OF TEMPERATURE THROUGH AN UNSTEADY EXPANSION WAVE IN PURE NITROGEN FOR VARIOUS EFFECTIVE VIBRATIONAL RELAXATION TIMES τ_s. (FROM REF. 13.)

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SPECTRUM-LINE-REVERSAL TEMPERATURE MEASUREMENTS THROUGH UNSTEADY RAREFACTION WAVES IN VIBRATIONALLY-RELAXING OXYGEN. Holbeche, T. A. June 1964. 546.21

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With oxygen initially at about 2500°K and 0.75 atmospheres pressure ($M_s \sim 6.9$, $p_1 \sim 10$ torr), the vibrational temperature was observed to fall from its initial (fully-frozen) value at the tail of the wave to a minimum before rising again to the original equilibrium value at the head of the wave.

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This agrees broadly with Appleton's calculations based on a linear rate law and Landau-Teller theory for τ_v, although the temperature minimum was slightly higher and occurred later than he predicted; it has also been predicted qualitatively by Stulov.

More detailed studies with, for example, nitrogen would be desirable for seeking a full explanation of the small differences with theory revealed by these preliminary tests.

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