Effect of Kinetic Rate Uncertainties on Gasdynamic Laser Gain and Energy Predictions

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Using an existing computer code, a study is made of the sensitivity of gasdynamic laser calculations to uncertainties in the vibrational energy exchange rates associated with CO_2–N_2–H_2O gas mixtures. The results show that calculations of gasdynamic laser gain and maximum available energy in the laser cavity can be subject to noticeable inaccuracy due to rate data uncertainties. Moreover, those rates to which the calculations are most sensitive are clearly delineated.

Introduction

THE first successful demonstration of a CO_2–N_2 gasdynamic laser took place in 1966, as described by Gerry.[1] Since then, the state-of-the-art has blossomed and matured at a rapid rate, as surveyed in Refs. 2–4. In particular, a number of gasdynamic laser experiments have been carried out in arc tunnels,[5-7] shock tunnels,[7-9] and combustion driven devices.[1,10-12] In many of these experimental investigations, comparisons have been made with theory for small-signal laser gain and/or laser power extraction. Fair agreement has been obtained in some but not all cases. With the rapid development of such lasers, there is an increasing need to obtain closer agreement between prediction and experiment; indeed, it is desirable to calculate gasdynamic laser performance as accurately as possible—ultimately to within a few percent.

At present, there is an inherent limitation to the accuracy with which such calculations can be made, namely, the uncertainty in the pertinent vibrational kinetic rates. The heart of a gasdynamic laser is the nonequilibrium nozzle expansion in which the population inversion is created, and the laser cavity downstream of the nozzle exit where power is extracted. In both these regions the computed nonequilibrium flow (and hence the theoretical laser performance) is dependent on a complex finite-rate vibrational energy exchange mechanism which is still not yet fully understood. For the CO_2–N_2–H_2O mixture common to most gasdynamic lasers, at least eight distinct vibrational kinetic rates must be utilized in the calculations. In turn, uncertainties in these rates will cause inaccuracies in the computed gasdynamic laser performance. Hence, two questions immediately arise: 1) what are the present uncertainties in the measured kinetic rates, and 2) how sensitive are calculations of gasdynamic laser performance to these uncertainties? These questions are addressed in the present investigation.

In particular, numerical experiments are carried out in order to test the sensitivity of gasdynamic laser gain and maximum available energy in the laser cavity to uncertainties in the kinetic rates. These calculations were made with an existing computer program described in Ref. 13. This computer code is based on the time-dependent nonequilibrium nozzle flow analysis discussed in Refs. 14 and 15, and applied to gasdynamic lasers in Ref. 16.

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Index categories: Lasers; Reactive Flows.

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Kinetic Rates

The vibrational kinetic rates pertinent to CO_2–N_2 gas lasers were examined and compiled by Taylor and Bitterman, first in 1967[17] and slightly updated in 1969.[18] These references are timely and excellent surveys, and have become a standard source for laser calculations. However, in 1969, Refs. 17 and 18 clearly show that a large scatter existed in much of the data; moreover, for a few rates, only one set of data were available. Additional CO_2–N_2–H_2O vibrational rates measured or calculated since 1969 are presented in Refs. 19 and 20, along with the standard Taylor and Bitterman data; this is an effort to assess uncertainties which currently exist in these rates.

The reactions that must be included in the calculation of small-signal gain and maximum available energy in gasdynamic lasers are as follows: 1) $CO_2^*(v_2)+H_2O \rightleftarrows CO_2+H_2O$; 2) $N_2^*+H_2O \rightleftarrows N_2+H_2O$; 3) $CO_2^*(v_3)+H_2O \rightleftarrows CO_2^{***}(v_2)+H_2O$; 4) $CO_2^*(v_2)+N_2 \rightleftarrows CO_2+N_2$; 5) $CO_2^*(v_2)+CO_2 \rightleftarrows 2CO_2$; 6) $CO_2^*(v_3)+N_2 \rightleftarrows CO_2^{***}(v_2)+N_2$; 7) $CO_2^*(v_3)+CO_2 \rightleftarrows CO_2^{***}(v_2)+CO_2$; and 8) $N_2^*+N_2 \rightleftarrows 2N_2$.

For such calculations, the pumping rate between excited N_2 and $CO_2(001)$, as well as the Fermi resonance between $CO_2(100)$ and $CO_2(020)$, are considered to be sufficiently rapid that local equilibrium is established between N_2 and $CO_2(v_3)$ as well as between $CO_2(v_1)$ and $CO_2(v_2)$. Hence, the rates for these reactions are not considered here. However, it should be noted that recent information has raised some questions about the local equilibrium normally assumed between the $CO_2(v_1)$ and $CO_2(v_2)$ modes. Specifically, Rosser, Hoag, and Gerry[21] present data which indicate that such equilibrium may not exist for all laser conditions. This contention is further supported by Bulthius,[22] who presents a measured rate for energy transfer from the (100)–(020) level which is an order of magnitude slower than previously reported values, and prompts a comment that the vibrational temperatures of the $CO_2(v_1)$ and $CO_2(v_2)$ modes are not always the same, particularly during peak power extraction. On the other hand, De Temple et al.[23] countered these contentions by presenting measurements for the decay rate of $CO_2(100)$ which are an order of magnitude faster than those of Rosser et al.[21] De Temple et al. feel that their measurements are more representative of the intrinsic V–V relaxation process, whereas those of Ref. 21 are more characteristic of a V–T process under the influence of a perturbing radiation field. More recently, Murray[24] also has measured different vibrational temperatures for $CO_2(v_1)$ and $CO_2(v_2)$. This matter, albeit an important one, is still in a state of flux; until it is resolved, the assumption of local equilibrium between the $CO_2(v_1)$ and $CO_2(v_2)$ modes appears to be the only logical recourse for gasdynamic laser calculations. Of course, if the vibrational temperature of $CO_2(v_1)$ were to lag that of

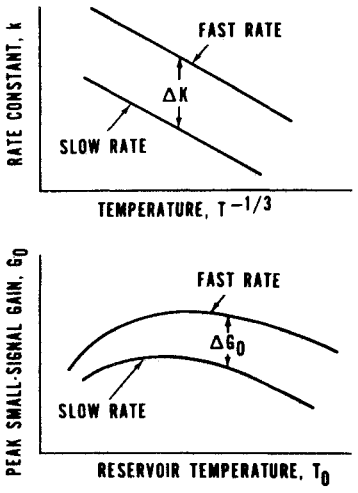

Fig. 1 Sketch defining parameters used in the numerical experiments.

$CO_2(v_2)$, then the resulting laser gain and power extraction would be decreased.

Numerical Experiments

The computer code described in Ref. 13 is used to calculate gasdynamic laser gain G_o and maximum available energy e_{max} as a function of reservoir temperature T_o. Results are obtained for both a typical first generation gasdynamic laser[2] (nozzle exit to throat area ratio $A_e/A^* = 20$, reservoir pressure $p_o = 20$ atm, throat height $h^* = 1$ mm, and mole fractions X_{CO_2} and X_{H_2O} equal to 0.07 and 0.01, respectively), and a second generation laser[2] ($A_e/A^* = 50$, $p_o = 37.5$ atm, $h^* = 0.365$ mm, $X_{CO_2} = 0.07$, $X_{H_2O} = 0.035$). Calculations were made alternately employing fast and slow rates for each kinetic reaction, one at a time. These rates are shown schematically at the top of Fig. 1. The vertical distance between the slow and fast rate constants k define a "change in rate constant" Δk. In turn, this change Δk causes a corresponding change in small-signal gain, ΔG_o, as shown at the bottom of Fig. 1. Hence, a *sensitivity factor* S can be defined for each rate as

$$S = (\Delta G_o/G_o)/(\Delta k/k)$$

Therefore, *S physically represents the uncertainty in gain per unit uncertainty in rate constant.* The actual values used for Δk along with the resulting ΔG_o for each kinetic reaction are detailed in Refs. 19 and 20, hence only the resulting sensitivity factors will be discussed here.

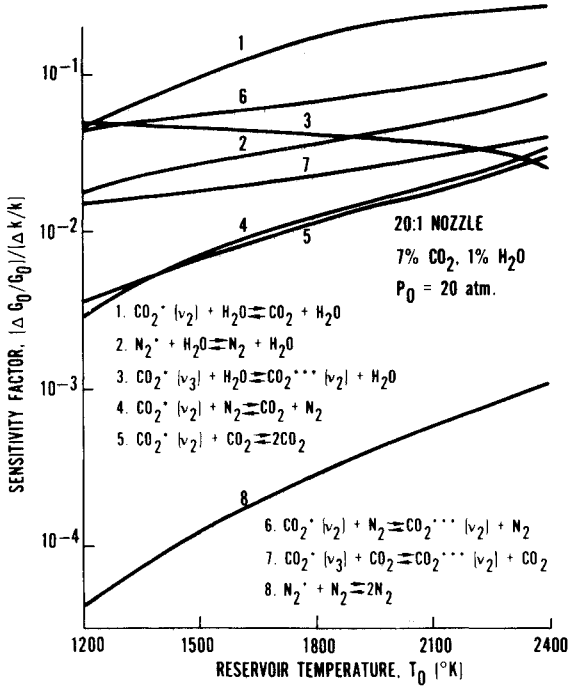

Fig. 2 Gain sensitivity factor; first generation gasdynamic laser.

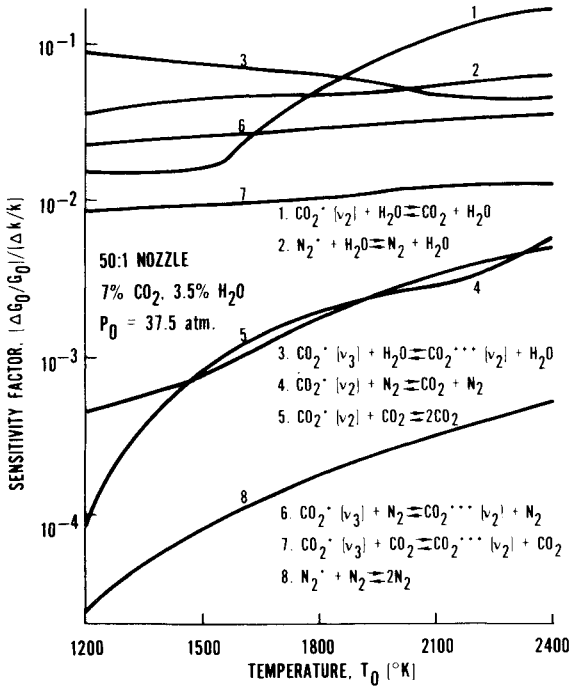

Fig. 3 Gain sensitivity factor; second generation gasdynamic laser.

Results

The sensitivity of gasdynamic laser gain to the various rates is clearly shown in Fig. 2 for a low nozzle area ratio case typical of first generation devices. Here, the sensitivity factor S is plotted as a function of reservoir temperature T_o; a curve is shown for each of the rates considered in the present investigation. For the case shown, the laser gain is most sensitive to reactions 1, 3, and 6, and least sensitive to reaction 8. In fact, for $T_o = 1200$K (which is typical of first generation gasdynamic lasers), the rather remarkable observation is made that the reactions

$$CO_2^*(v_2) + H_2O \rightleftarrows CO_2 + H_2O$$
$$CO_2^*(v_3) + H_2O \rightleftarrows CO_2^{***}(v_2) + H_2O$$
$$CO_2^*(v_3) + N_2 \rightleftarrows CO_2^{***}(v_2) + N_2$$

all exert equal and maximum sensitivity on calculations of gasdynamic laser gain.

Similar results are shown in Fig. 3 for a case typical of high area ratio, high H_2O content, second generation devices. Here, in addition to reactions 1, 3, and 6, reaction 2 is seen to be important. In fact, at $T_o = 1800$K (which is typical of second generation devices), the reactions

$$CO_2^*(v_2) + H_2O \rightleftarrows CO_2 + H_2O$$
$$N_2^* + H_2O \rightleftarrows N_2 + H_2O$$
$$CO_2^*(v_3) + H_2O \rightleftarrows CO_2^{***}(v_2) + H_2O$$

exert the most sensitivity on calculations of gasdynamic laser gain.

Reflecting on Fig. 3, emphasis is made of the importance of the reaction $N_2^* + H_2O \rightleftarrows N_2 + H_2O$. In contrast, Schneider and Holmes[25] draw the conclusion that gain and power are only slightly sensitive to the rate for deactivation of N_2 by H_2O.

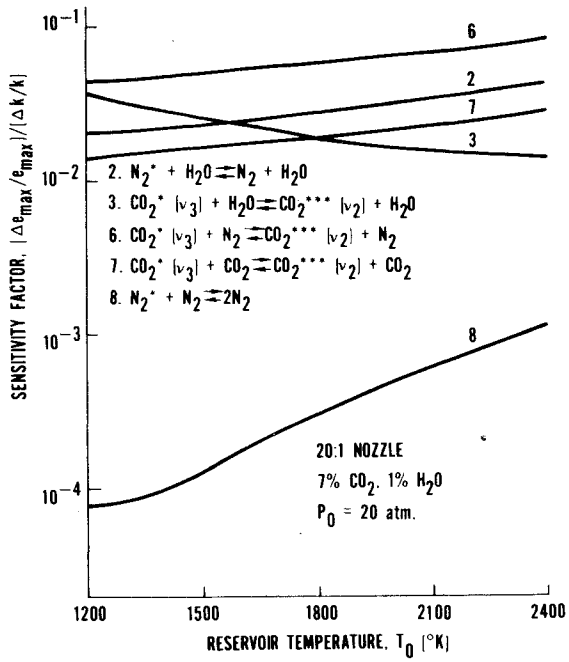

Fig. 4 Maximum available energy sensitivity factor; first generation gasdynamic laser.

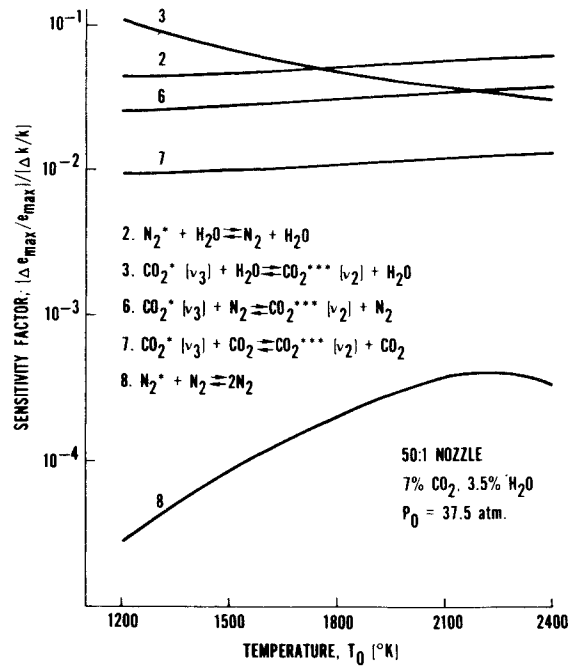

Fig. 5 Maximum available energy sensitivity factor; second generation gasdynamic laser.

However, different mixtures at different conditions are used, so no direct comparison can be made. The 3.5% H_2O content used in Fig. 3 will certainly result in more sensitivity than the 1% content used in Ref. 25. However, referring again to Fig. 2, which is for a 1% H_2O content, reaction 2, even though not among the most sensitive, still can not be totally discounted. Additional insight can be gained by examining the magnitude of the rate constant for reaction 2. At a temperature of 1400K, which is typical of the static temperature of the gas in the throat region of gasdynamic laser nozzles (where most of the vibrational freezing is occurring), k is approximately 10^{-12} cm^3/sec for reaction 2. This is comparable to the rate of 2×10^{-12} cm^3/sec for reactions 1 and 3, the fastest of any of reactions 1–8. This can be seen from the data compiled in Refs. 19 and 20. Hence, simply because of its fast rate, reaction 2 should exert a noticeable influence on gasdynamic laser calculations. This question is worth future study.

Considering maximum available energy e_{max} a sensitivity factor can also be defined analogous to that for gain. This factor is shown in Figs. 4 and 5 for the first and second generation laser cases, respectively. Curves for the rates involving deactivation of the lower laser level are not shown because they have a minimal effect on e_{max}. Comparing Figs. 4 and 5 with Figs. 2 and 3, the reactions that exert maximum and minimum sensitivities on e_{max} are the same that exert maximum and minimum sensitivity on G_o.

The results shown in Figs. 2–5 are uncertainties in performance *per unit uncertainty in rate constant*, hence they indicate the *sensitivity* of gasdynamic laser calculations to the individual rates. However, some rate constants are known more accurately than others; hence, the question is now posed, what are the absolute uncertainties in G_o and e_{max} due to the existing absolute uncertainties in rate constants? Some absolute uncertainties are given in Table 1, and were obtained from examination of the data scatter shown in Refs. 19 and 20.

These uncertainties are subjective judgments. They are obtained in Refs. 19 and 20 by comparing old and new data from the existing literature, with no particular knowledge of which data may be more valid than the other. Hence, the foregoing values of $\Delta k/k$ are probably conservative and do not represent the real uncertainties which exist today. The real uncertainties are probably less than shown above. Nevertheless, they are useful to establish some upper bounds.

Table 1 Absolute uncertainties in rate constants.

Reaction	Uncertainty in data, $\Delta k/k$
1) $CO_2^*(v_2)+H_2O \rightleftarrows CO_2+H_2O$	3
2) $N_2^*+H_2O \rightleftarrows N_2+H_2O$	5
3) $CO_2^*(v_3)+H_2O \rightleftarrows CO_2^{***}(v_2)+H_2O$	5
4) $CO_2^*(v_2)+N_2 \rightleftarrows CO_2+N_2$	4
5) $CO_2^*(v_2)+CO_2 \rightleftarrows 2CO_2$	0.5
6) $CO_2^*(v_3)+N_2 \rightleftarrows CO_2^{***}(v_2)+N_2$	5.0
7) $CO_2^*(v_3)+CO_2 \rightleftarrows CO_2^{***}(v_2)+CO_2$	0.25
8) $N_2^*+N_2 \rightleftarrows 2N_2$	0.5

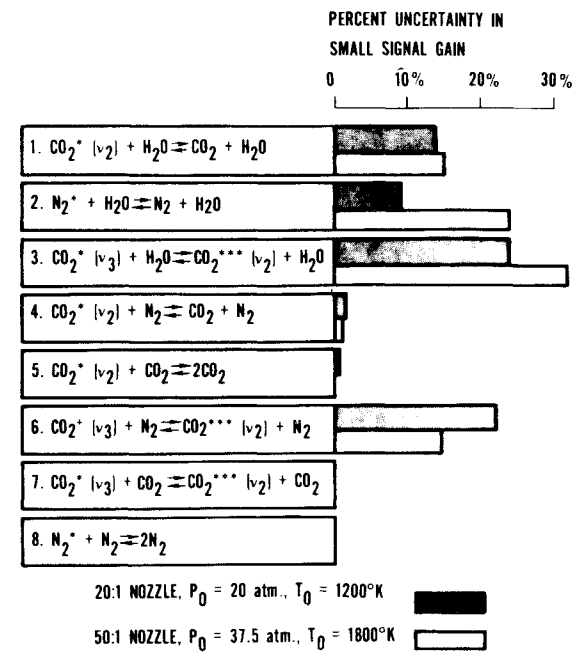

Fig. 6 Some upper bounds on uncertainty in small-signal gain.

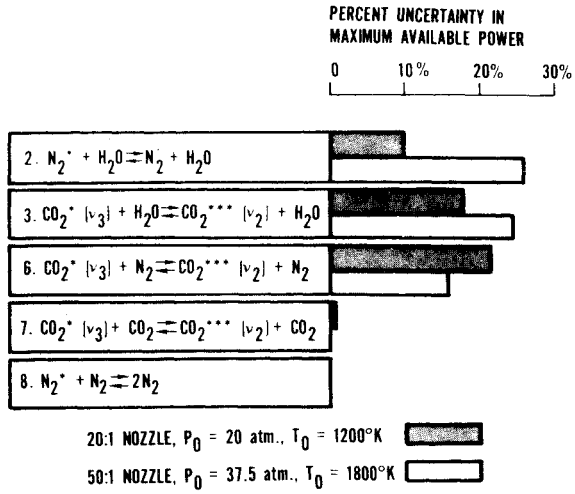

Fig. 7 Some upper bounds on uncertainty in maximum available energy in the laser cavity.

When the sensitivity factors shown in Figs. 2–5 are multiplied by the foregoing absolute rate data uncertainties $\Delta k/k$, over-all uncertainties in G_o and e_{max} are obtained. These results are shown as bar charts in Figs. 6 and 7 for G_o and e_{max}, respectively. In Figs. 6 and 7, reservoir temperatures of 1200K and 1800K are chosen as typical of first and second generation gasdynamic lasers, respectively. Note from Figs. 6 and 7 that the maximum absolute uncertainties are on the order of 30%. *The reader should construe this only as some measure of an upper bound; as previously discussed, the true uncertainty is most likely smaller.*

In the foregoing results, the kinetic rates were changed one at a time in order to isolate their individual influence on the calculations. In order to examine the combined effect of rate uncertainties two additional calculations are made wherein, first, all slow rates are used simultaneously, and alternately, all fast rates are used. The fast and slow rates for these calculations were obtained from the absolute uncertainties $\Delta k/k$ listed above, centered around a mean reference value given by the reference rates (or relaxation times) quoted in Appendix A. The results are given in Fig. 8 for small-signal gain as a function of T_o. Again, these results should be interpreted as very conservative bounds; the real uncertainty is probably much less.

Conclusions

From the present study of the sensitivity of gasdynamic laser gain and maximum available energy to uncertainties in the vibrational rate data, two conclusions are drawn.

1) The calculated results are most sensitive to the following processes:

$$CO_2^*(v_2) + H_2O \rightleftarrows CO_2 + H_2O$$
$$CO_2^*(v_3) + H_2O \rightleftarrows CO_2^{***}(v_2) + H_2O$$
$$CO_2^*(v_3) + N_2 \rightleftarrows CO_2^{***}(v_2) + N_2$$
$$N_2^* + H_2O \rightleftarrows N_2 + H_2O$$

It is important that accurate rates, along with their proper temperature dependence, be utilized for these reactions.

2) At most, gasdynamic laser gain and maximum available energy are uncertain to about 30% because of the major uncertainties between old and new rate data in the existing literature. In reality, using some insight as to which rates are more valid, the actual uncertainty in gasdynamic laser calculations could be much less.

Appendix A

The reference rates used to generate most of the data in this paper are given below. They are the same as appear in Refs. 19 and 20. The nomenclature is the same as Refs. 13 and 16.

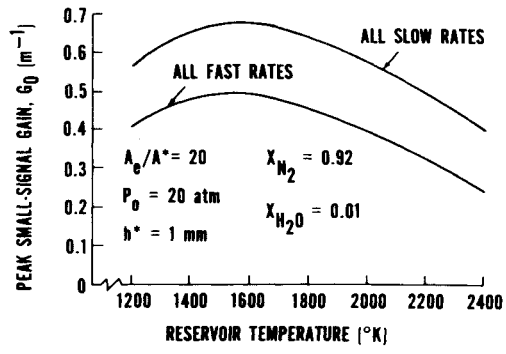

Fig. 8 Uncertainties in gain bounded by simultaneously all fast rates and simultaneously all slow rates.

$$(\tau_a p)_{CO_2\ N_2} = 1.3 \times 10^5 (T^{-1/3})^{4.9}$$
$$(\tau_a p)_{CO_2\text{-}CO_2} = 0.27(\tau_a p)_{CO_2\ N_2}$$
$$(\tau_a p)_{CO_2\text{-}H_2O} = 5.5 \times 10^{-2}$$
$$\log(\tau_b p)_{N_2\ N_2} = 93(T^{-1/3}) - 4.61$$
$$(\tau_b p)_{N_2\text{-}CO_2} = (\tau_b p)_{N_2\text{-}N_2}$$
$$\log(\tau_b p)_{N_2\ H_2O} = 27.65(T^{-1/3}) - 3.2415$$
$$\log(\tau_c p)_{CO_2\ CO_2} = 17.8(T^{-1/3}) - 1.808$$
$$(\tau_c p)_{CO_2\text{-}N_2} = 2(\tau_c p)_{CO_2\ CO_2}$$
$$\log(\tau_c p)_{CO_2\ H_2O} = -20.4(T^{-1/3}) + 0.643 \quad (\text{For } T > 600\text{K})$$
$$\log(\tau_c p)_{CO_2\ H_2O} = -20.4(600^{-1/3}) + 0.642 = \text{constant} \quad (\text{For } 200 < T < 600\text{K})$$

In the previous correlations, (τp) is in (μsec-atm) and T is in degrees K.

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Blast Initiation and Propagation of Cylindrical Detonations in MAPP-Air Mixtures

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An experimental investigation of the initiation, transition and quasi-steady propagation of blast initiated cylindrical detonations in Methyl Acetylene, Propane, Prodadiene (MAPP)-air mixtures is described. A sectored shock tube was employed which was originally designed to allow study of cylindrical shock, and homogeneous and heterogeneous detonation waves. Cylindrical blast waves were generated by firing of controlled amounts of a condensed explosive at the apex of the sector. Experimental data served to suggest the existence of three wave propagation regimes: subcritical energy regime, where decoupling of shock and reaction zone results in a reacting blast wave-type decay; the critical energy regime, where decoupling occurs but is followed by the re-establishment of a sub-Chapman-Jouguet condition, with an asymptotic strengthening to the CJ state; and supercritical energy regime, where the initially overdriven detonation decays asymptotically to its CJ state. Threshold energy levels delineating the subcritical to critical energy regimes were established for a wide range of MAPP-air mixtures. Lean and rich limits for steady propagating detonation waves were also established. Comparisons made with the work of others reveal that these limits do not suffer from scale effects. The detonation velocity, when attained, displayed a definite dependence on blast wave energy with the higher energy runs giving reasonable agreement with theory. The measured transition distances from blast to detonation wave, nondimensionalized by the blast wave explosion length, compared satisfactorily with theoretical predictions.

Introduction

In recent years much concern has been generated regarding the frequent occurrence of accidental ignition and subsequent explosion of large unconfined vapor clouds. Explosions are commonly thought of as occurring in a confined environment such as in vessels, pipes, sewers, homes, and the like. On the other hand, it has become increasingly evident that unconfined explosions may be generated in the atmosphere. As pointed out by Strehlow,[1] the explosion of unconfined vapor clouds by the dispersion of flammable liquid or vapor spills is becoming a serious problem, mainly because of the increased size of these spills. Further, there has been interest in delibrate fuel-air explosions wherein a liquid of gaseous fuel is dispersed throughout a volume of the atmosphere and detonated. To date, quantitative experimental studies of unconfined explosions are essentially nonexistent and therefore current practice of damage assessment and/or risk evaluation is based on the estimate of a TNT equivalent from the damage patterns, assuming a point source blast wave energy release. It is known, however, that unless the cloud detonates, the explosion itself and the blast wave it produces are far from ideal and cannot be approximated adequately by classical self-similar solutions. Theory substantiated by experiment has succeeded in classifying the attendant wave phenomena into energy-dependent regimes. In any case, the exact energy release rate is of fundamental importance and is very dependent on initial conditions, such as spill geometry, fuel distribution, turbulence and buoyancy effects, and the type and energy of the ignition source.

Because of the complex nature of unconfined fuel-air explosions, a detailed analytic solution is not completely

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