Investigation on the Theoretical Modeling of a Supersonic Flow CO Chemical Laser*

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Small signal gain coefficients of a supersonic flow CO chemical laser are calculated using a theoretical model which was developed previously and compared with the corresponding experimental results. It is shown that the model predicts the gain characteristics only qualitatively. The measured small signal gain coefficients are rather small compared with the calculated results. In the present investigation, the theoretical model is improved significantly by modifying the relaxation rate constants and by including the effects of the boundary layers which develop in the optical test section. However, further improvement on the theoretical model is necessary by including the effects of the nozzle boundary layers and shock waves.

Key Words: Fluid Mechanics, Supersonic Flow, Laser, Carbon Monoxide, Chemical Laser, Reactive flow

1. Introduction

The CO chemical laser⁽¹⁾ is of continuing interest because of the high efficiency of chemical pumping, the high quantum efficiency and the slow vibrational relaxation. However, the supersonic flow CO chemical laser has received relatively limited experimental attention⁽²⁾⁽³⁾ and the details of its performance characteristics are not sufficiently known. In the previous papers⁽⁴⁾⁽⁵⁾, the authors developed a theoretical model of a supersonic flow CO chemical laser. A shock tunnel facility was also fabricated and the gain characteristics were studied experimentally⁽³⁾. In the present paper, the propriety of the theoretical model developed previously is examined using the experimental data. The chemical luminescence from the various reactions in the laser cavity is also observed in

order to improve the theoretical model.

2. Outline of Experiment

In the previous experiments⁽³⁾, high-temperature mixtures of CS₂, CS, S₂, S and Ar were produced in a shock tube where the thermal dissociation of CS₂ diluted in Ar was accomplished by a reflected shock wave. The shock-heated mixture was exhausted through supersonic nozzles which froze the recombination kinetics, thereby preserving the high CS and S concentrations, and generated the low-temperature supersonic streams. Downstream of the nozzle exits, the chemically nonequilibrium streams were mixed with supersonic streams of O₂ and the chain reaction

began to form the vibrationally excited CO molecules.

Figure 1 shows the supersonic nozzles and the optical test section in which x is the direction of the flow with y parallel to the optical axis and z is the direction perpendicular to these axes. Three sets of CS₂ nozzles and O₂ nozzles were used in the present experiments. The nozzles were designed as wedge nozzles. The throat height, the area ratio and the

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Fig. 1 Supersonic nozzles and the optical test section

angle of divergence of the CS_2 nozzle were 0.6 mm, 41.3, and 40° , respectively, and those of the O_2 nozzle were 0.5 mm, 10, and 60° , respectively. The channel downstream of the nozzle bank had a constant cross-sectional area of $24 \text{ mm} \times 90 \text{ mm}$. Measurements of small signal gain coefficients were made using four pairs of circular windows, as shown in Fig. 1. The details of the shock tube, the O_2 supply system and the method of the experiment are shown in the previous paper⁽³⁾.

3. Outline of Theoretical Model

Figure 2 shows a schematic plane view of the flow field. A leaky stream tube approach⁽⁵⁾ is applied to the flow field downstream of the nozzle exits. The flow field is divided into three stream tubes, the first stream tube for the dissociated products of CS_2 diluted in Ar, the second for O_2 , with the third one located between these two. The mixing occurs in the third stream tube. In Fig. 2, p , s and m are used to designate the stream of the dissociated products of CS_2 , the O_2 stream and the mixed stream, respectively. It is assumed that each stream is quasi-one-dimensional and the static pressure is uniform across all stream tubes. It is also assumed that the m stream is fully developed turbulent shear layers. Therefore, the width Y_m of the m stream is written as follows:

$$Y_m = \delta_T x. \quad (1)$$

In principle, the coefficient δ_T could be determined by fitting the experimental observations, for example, of the growth of the chemiluminescent part of the mixing layer. Varwig⁽⁶⁾ and Driscoll et al.⁽⁷⁾ showed by photographic observations of the HF and DF chemical laser reacting flow fields that the value of δ_T is between 0.1 and 0.03. The details of the theoretical model are shown in the previous papers⁽⁴⁾⁽⁵⁾.

Fig. 2 Schematic of flow regions

4. Results

In the previous experiments, the initial mole fractions of CS_2 and Ar in the driven section of the shock tube were 0.1 and 0.9 respectively. The plenum of the O_2 was room temperature. For a given plenum temperature T_{0p} and plenum pressure P_{0p} of the p nozzle, the plenum pressure P_{0s} of the s nozzle was regulated so that the pressure at the exit of the s nozzle agrees with that of the p nozzle. A method⁽⁴⁾ previously developed was used to calculate P_{0s} .

Figures 3, 4 and 5 show the dependence of the calculated small signal gain coefficients G for $P_8(12)$, $P_8(12)$ and $P_{10}(12)$ transitions on distance x from the nozzle exit. These results are obtained assuming $\delta_T = \infty$ (instantaneous mixing at the nozzle exit $x=0$), 0.1 and 0.03 respectively. Figure 6 shows the experimental results⁽³⁾ obtained previously. It is noted that the scales of Fig. 6 are different from those of Figs. 3, 4 and 5. The calculated distributions obtained assuming different values of δ_T agree with each other qualitatively. As the dissociated CS_2 streams mix with the O_2 streams, gains of all branches grow gradually due to the formation of the vibrationally excited CO molecules. However, the gains arrive at the peaks, then decay rapidly to negative values due to the vibrational relaxation.

Thereafter, they grow again and approach values (almost zero) attained at the Boltzmann distribution. The existence of large absorption can be easily under-

Fig. 3 Predicted distributions of the small signal gain coefficients along the flow ($\delta_T = \infty$)

Fig. 4 Predicted distributions of the small signal gain coefficients along the flow ($\delta_T = 0.1$)

Fig. 5 Predicted distributions of the small signal gain coefficients along the flow ($\delta_T = 0.03$)

stood using the vibrational population distribution of the CO molecules. In Fig. 5, A, B, C, and D are used to designate the locations where the small signal gain coefficient of the $P_8(12)$ branch is maximum, zero, minimum, and almost the final value, respectively. The vibrational population distributions of CO at these locations are shown in Fig. 7, where $n_{co,v}$ is the fraction of CO at vibrational level v . Powell⁽⁸⁾ showed that the reaction (2) produces CO with a non-Boltzmann population distribution among the vibrational energy levels with maximum population at $v=13$, as shown in Fig. 7. At location A, the populations at $v=7$ and 8 are inverted considerably. However, the distribution evolves by continued formation and by rearrangement through VV and VT collisions. The

Fig. 6 Measured distributions of the small signal gain coefficients along the flow

Fig. 7 Dependence of $n_{co,v}$ on v

optimal ν shifts to lower values in the downstream direction. As a result, the difference between the populations at these levels almost disappears at location B. The population at $\nu=7$ surpasses that at $\nu=8$ significantly at location C. Further downstream, at location D, the distribution approaches the Boltzmann distribution, as shown in Fig. 7. Since the populations at $\nu=7$ and 8 are very small, the difference between the populations is also very small. The measured small signal gain coefficients shown in Fig. 6 also increase downstream of the nozzle exits and then begin to decrease after they arrive at the maximum values. In accordance with the theoretical results, the growth and decay are faster for gains of $P_\nu(J)$ branches with large ν values. In the previous experiments, large absorption could not be observed since there were no windows at $x \approx 0.2$ m. However, it seems that the data on the $P_{10}(12)$ branch suggest the existence of large absorption. Therefore, the existence of large absorption is shown in Fig. 6 using the dashed lines. It can be clearly seen that the theoretical results obtained when assuming $\delta_T = \infty, 0.1$, and 0.03 , considerably overestimates the small signal gain coefficients and the active region where positive gains can be preserved, although it can qualitatively predict the distributions of the small signal gain coefficients well. The maximum gain can be decreased if a smaller δ_T is assumed. However, the smaller δ_T leads to the larger active region. Therefore, the predicted distribution cannot be adjusted to those of the experiments by the proper choice of δ_T . It is thought that the present model considerably overestimates the formation of the excited CO molecules and/or underestimates the vibrational relaxation.

5. Discussion

The theoretical model of the supersonic flow CO chemical laser consists of chemical reaction, vibrational relaxation, and gas dynamics. However, the model of the chemical reactions⁽⁴⁾ is reasonably reliable since many experiments on CS₂ flames have been made, and a large body of experimentally determined information has resulted. In what follows, the theoretical models of vibrational relaxation and gas dynamics are reexamined.

5.1 Vibrational relaxation

The reexamination of the vibrational kinetic model developed previously⁽⁴⁾ reveals the following issues.

i) The most important relaxation process in the CO chemical laser is the redistribution of the vibrational energy in the chemically pumped CO by the intramolecular VV transfer :

In the previous paper⁽⁴⁾, the rate constant K was calculated for all the vibrational relaxation processes using the SSH theory⁽⁹⁾ and modified by the experimental data if available. On the other hand, Rookwood et al.⁽¹⁰⁾ proposed the kinetic rate constants for general process (4) by calculating the transition probabilities based on the short-range force and the long-range force. The rate constants for process (4) proposed by the present authors⁽¹⁰⁾ and Rookwood et al.⁽¹⁰⁾ are shown in Fig. 8 and compared with those of the experimental data⁽¹⁰⁾⁽¹³⁾⁽¹⁴⁾. The rate constants calculated using the SSH theory, which is based only on the short-range force, agree with the experiments for small ν only. However, the rate constants proposed by Rookwood et al. agree very well with the experiments even for large ν . Therefore, it is desirable to apply Rookwood's rates to process (4).

ii) The other VV and VT transfers between the diatomic molecules involved in the previous model are reexamined using a method suggested by Shin⁽¹¹⁾ which treats the potential functions with more precision than the SSH theory. It is shown that the rate constants calculated by Shin's theory are considerably higher than those by the SSH theory for the VT transfer :

where M is the collision partner. In the previous model⁽⁴⁾, the collisions with species other than Ar and O, are neglected for processes (5). However, Shin's theory suggests that the rate constants of process (5)

Fig. 8 The rate constants for vibrational transition (4)

for collision partners CS, S₂, SO and CO are nearly equal to those for O₂.

iii) Shin's theory is applied to processes which were neglected in the previous model. It is revealed that the rate constants are relatively large for the following processes:

Based on the reexamination, the gain characteristics are calculated using the following four models:

A: previous model⁽⁴⁾

B: model refined by i)

C: model refined by i) and ii)

D: model refined by i), ii) and iii).

The calculated gain profiles are shown in Fig.9 and compared with the experimental data. The refinement of the vibrational kinetic model leads to faster growth, faster decay and a lower maximum value of the small signal gain coefficient. However, the agreement with the experiments is still not satisfactory. It is inferred that the discrepancy between the theoretical results and the experiments is mostly due to deficiency in modeling the gas dynamics. In the following calculations, vibrational kinetic model C is used since the results obtained using models C and D almost coincide with each other.

5.2 Gas dynamics

The small signal gain coefficients depend very strongly on the temperature T , since the vibrational rate constants⁽⁹⁾ are nearly proportional to $\exp(T^{-1/3})$. Therefore, the temperature calculated using the pres-

Fig. 9 Predicted distributions of the small signal gain coefficient along the flow using various vibrational kinetic models

ent gas dynamics model might be considerably lower than that in the actual flow. The temperature is estimated to be lower if the effects of boundary layers are neglected. In the present study, therefore, the visual observation of the cavity flow field is also conducted utilizing the chemical luminescence from various reactions. Figure 10 shows a typical photograph of the chemiluminescence which is taken using the circular windows shown in Fig.1. The photograph clearly shows that the strong white-blue chemiluminescence exists in the central portion of the laser cavity due to the recombination of CS and S into CS₂. However, it is also observed that reddish layers develop along the upper and lower walls of the cavity and the region of white-blue chemiluminescence almost disappears at the third window. The authors suspect that the reddish layers are boundary layers. It is noted that the measurements⁽³⁾ of the small signal gain coefficients were made in the central portion of the cavity using a diagnostic laser beam of 3 mm in diameter. The flow fields near the upper and lower walls are highly three-dimensional. Therefore, it is not easy to analyse the cavity wall boundary layers. We assume, however, that only the p streams issuing from the nozzles exist in the cavity. This assumption is due to the fact that the p streams occupy the greater part of the flow field of interest if $\delta^* = 0.1$ or 0.03 is assumed. We also assume that the cavity boundary layers develop from the nozzle exits. The Reynolds number which determines the turbulence transition is written as follows:

$$Re_x = \rho_\infty u_\infty x / \mu, \quad (12)$$

where ρ is the density, μ is the viscosity and the subscript ∞ represents the edge conditions. It can be assumed that the cavity boundary layers are laminar, since the Reynolds number based on the edge conditions and $x = 0.25$ m (the location of the third window) is only 2.6×10^5 . For compressible boundary layers along a flat plate, the displacement thickness δ^* can be calculated using the following equation⁽¹²⁾:

$$\delta^*/x = Re_x^{-1/2} [1.73 T_w/T_0 + 0.332(\gamma-1)M_\infty^2], \quad (13)$$

where T_0 is the plenum temperature, T_w is the wall

Fig. 10 Chemical luminescence from the cavity and the calculated boundary displacement thickness

Fig. 11 The effects of boundary layers on the distribution of the small signal gain coefficient

temperature, γ is the specific heat ratio and M_∞ is the edge Mach number. Assuming that $T_w = 300$ K and that the edge conditions are equal to the exit conditions of the p nozzle, δ^* is calculated and shown in Fig.10. Since the boundary layer thickness is several times as large as the displacement thickness, the reddish layers observed in the photograph must be boundary layers. The presence of the boundary layers causes the convergence of the effective cross sectional area of the cavity. In the present study, the cross sectional area of the cavity is modified by δ^* and the gain characteristics are calculated. The results are shown in Fig.11 and compared with those of the experiments. It is seen that the maximum value of G is increased slightly due to the presence of the boundary layers since the number densities of reactants are increased. However, the presence of boundary layers leads to considerably faster growth and faster decay of G , since the temperature is raised considerably. Although the refinement of the model is very simple, it clearly demonstrates that the effects of boundary layers cannot be neglected. However, the agreement of the theoretical results with the experiments is still rather unsatisfactory. One of the reasons is that the estimation of the boundary layers is not very precise. In order to evaluate them precisely, the following phenomena must be considered.

1) Downstream of the second window, the greater part of the flow field is occupied by the boundary layers, as shown in the photograph. Therefore, it is desirable to consider the effects of viscosity in the whole flow field.

2) The temperature is raised and the velocity is made slower in a flow where the heat due to the chemical reaction is produced even if the cross-sectional area remains constant. The presence of bound-

ary layers also elevates the temperature and lowers the velocity. Therefore, the boundary layers might develop faster and transition to turbulence might occur.

3) The actual boundary layers are highly three-dimensional.

4) The chemical reactions and the vibrational relaxations should be considered in the calculation of the boundary layer.

The reasons other than the effects of boundary layers for disagreement between the theoretical results and the experimental results are as follows.

1) The oblique shock waves generated at the trailing edge of the nozzle are neglected in the theoretical model.

2) The boundary layers which develop along the nozzle walls are neglected in the theoretical model.

It is clear that further refinement of the theoretical model is necessary in order to predict precisely the gain characteristics of the supersonic flow CO chemical laser.

6. Conclusions

The following conclusions can be drawn from the present investigation on the supersonic flow CO chemical laser.

1) The theoretical model which was developed previously⁽⁴⁾⁽⁵⁾ predicts the gain characteristics only qualitatively. The measured small signal gain coefficients are rather small compared with the calculated results.

2) Although the theoretical model can be refined by correcting the rate constants for VV and VT transfers of CO, the accuracy of the prediction is still not satisfactory.

3) The theoretical model is improved considerably by including the effects of boundary layers which develop along the cavity wall. However, further improvement on the theoretical model is necessary by refining the treatments of the cavity boundary layers and by including the effects of nozzle boundary layers and shock waves.

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