Time-Dependent Analysis of an N_2O Gasdynamic Laser

K. P. J. Reddy*

Indian Institute of Science, Bangalore, India

A performance study of an N_2O-N_2-He gasdynamic laser (GDL) is presented. A simplified two-temperature model is proposed for the vibrational energy levels of the N_2O and N_2 molecules. The governing equations of unsteady quasi-one-dimensional flow of N_2O, N_2, and He mixture in a convergent-divergent contour nozzle are solved using a time-dependent numerical technique. The final steady-state distributions of vibrational temperatures, population inversion, and the small-signal laser gain along the nozzle are presented. A highest value of 0.675 m^{-1} small-signal gain on the $P(19)(001) \rightarrow (100)$ transition at 10.9 μm of N_2O is obtained for a plenum temperature of 1200 K, plenum pressure of 6.9 atm, and the gas mixture composition N_2O:N_2:He = 25:45:30 (%). It is shown that, for plenum temperatures below 1200 K, an N_2O GDL is more efficient than the CO_2 GDL for identical operating conditions.

Nomenclature

A	= local cross-sectional area of the nozzle
C_{N_2O}, C_{N_2}	= mass fractions of N_2O and N_2
e	= specific energy
$(e_{vib})_I, (e_{vib})_{II}$	= vibrational energies of modes I and II
G_0	= small-signal laser gain
M_i	= molecular weight of species $i, i = N_2O, N_2$, He
N_{100}, N_{001}	= populations in (100) and (001) levels of N_2O
N_{N_2O}	= local number density of N_2O molecules
p	= pressure
p_0	= plenum pressure
R	= mixture gas constant per unit mass
R_{N_2O}, R_{N_2}	= specific gas constants of N_2O and N_2
t	= time
T	= translational temperature
T_0	= plenum temperature
T_I, T_{II}	= vibrational temperatures of modes I and II
u	= velocity of the laser gas mixture
x	= distance along the nozzle in the flow direction
X_i	= mole fractions of species $i, i = N_2O, N_2$, He
Z	= collision frequency
$\theta_1, \theta_2, \theta_3$	= characteristic vibrational temperatures of N_2O
θ_N	= characteristic vibrational temperature of N_2
λ	= laser wavelength
ν_1, ν_2, ν_3	= vibrational mode frequencies of N_2O
ν_N	= vibrational frequency of N_2
ρ	= mixture gas density
$\sigma_{N_2O\text{-}i}$	= collision cross sections of N_2O with species i
τ_I, τ_{II}	= characteristic average relaxation time of modes I and II
τ_{12}	= spontaneous radiative lifetime
τ_a, τ_b, τ_c	= relaxation times defined in Eqs. (15–17)

Introduction

BECAUSE the nitrous oxide (N_2O) molecule is in many respects similar to the CO_2 molecule, attempts have been made to generate laser radiation using N_2O gas. The first electrically excited N_2O laser was reported in 1965.[1] However, N_2O electric-discharge lasers are found to be inferior to CO_2 lasers in small-signal gain and power output. The inferior performance of the N_2O laser is due to the appreciable dissociation of N_2O molecules during collisions with the electrons in the discharge. In addition, it has been shown recently that electron de-excitation severely limits the population inversion that can be established in the upper laser level (00°1) and is the primary factor accounting for the poor performance of the N_2O laser relative to the CO_2 laser.[2]

However, because the N_2O laser has certain advantages for long-range communications and also has twice as many lines covering approximately the same spectral region as the CO_2 laser, efforts are made to improve the performance of the N_2O laser. The major limiting factors found in the electric-discharge N_2O laser can be reduced to a minimum in a gasdynamic regime, where the population inversions are produced by purely thermal means. Therefore, there has been some recent effort to study the performance of an N_2O gasdynamic laser (GDL) both theoretically and experimentally.[3-6] All of these studies indicate the effectiveness of the N_2O molecules as the working medium of a GDL.

In this paper, we report the theoretical analysis of the performance of an N_2O-N_2-He GDL. Unlike the earlier theoretical studies, the present analysis utilizes the unsteady fluid dynamic equations for the supersonic expansion of a mixture of N_2O, N_2, and He through a supersonic nozzle. In addition, a simplified three-temperature model, which simplifies the complex vibrational equations for the N_2O-N_2 system, is proposed and used here. The unsteady quasi-one-dimensional flow equations, augmented by a set of vibrational relaxation equations, are solved by using a time-dependent technique to obtain flowfield parameters. Steady-state values are approached asymptotically at large times by this technique. These steady-state solutions are further used to obtain the population inversion and the small-signal gain of the laser along the nozzle.

The effects of the dissociation of the N_2O molecules at high temperatures on the laser performance are neglected in this analysis.

Vibrational Kinetics

The linear asymmetric N_2O molecule is, in many respects, similar to the CO_2 molecule. Like the CO_2 molecule, the N_2O molecule has three normal modes of vibration: a symmetric
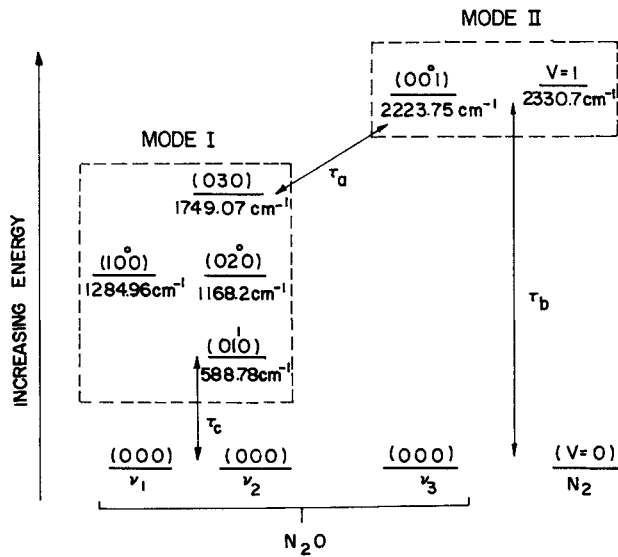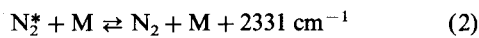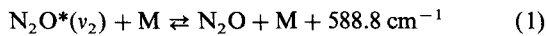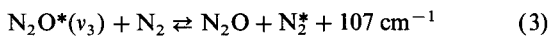
Fig. 1 Schematic of the simplified vibrational kinetic model for the N_2O-N_2 system.

mode with $\nu_1 = 1285$ cm^{-1}, a doubly degenerate deformation mode with $\nu_2 = 588.8$ cm^{-1}, and an asymmetric mode with $\nu_3 = 2223.5$ cm^{-1}. The laser transition $(00°1) \rightarrow (10°0)$ produces infrared radiation near 10.9 μm. The energy transfer processes that determine the vibrational energy balance in the N_2O-N_2-He system can be written in the form of detailed kinetic reactions as follows:
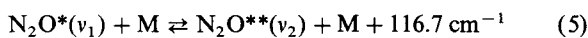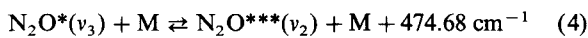
T-V processes:

$$N_2O^*(\nu_2) + M \rightleftarrows N_2O + M + 588.8 \text{ cm}^{-1} \qquad (1)$$

$$N_2^* + M \rightleftarrows N_2 + M + 2331 \text{ cm}^{-1} \qquad (2)$$

V-V processes (intermolecular):

$$N_2O^*(\nu_3) + N_2 \rightleftarrows N_2O + N_2^* + 107 \text{ cm}^{-1} \qquad (3)$$

V-V processes (intramolecular):

$$N_2O^*(\nu_3) + M \rightleftarrows N_2O^{***}(\nu_2) + M + 474.68 \text{ cm}^{-1} \qquad (4)$$

$$N_2O^*(\nu_1) + M \rightleftarrows N_2O^{**}(\nu_2) + M + 116.7 \text{ cm}^{-1} \qquad (5)$$

In the preceding equations, M stands for the collision partner that can be N_2O, N_2, or He, and the asterisks indicate the vibrational level in a given mode. We note the following pertinent observations of the N_2O-N_2 system.

1) The first excited level (100) of the ν_1 mode and the second excited level (020) of the ν_2 mode are relatively close, as shown by Eq. (5). Recently, it has been experimentally shown that ν_1 and ν_2 modes of N_2O are strongly coupled and have the same vibrational temperatures.[2] Hence, this reaction is extremely fast.

2) The energy difference between the first excited level (001) of the ν_3 mode of N_2O and the first vibrational level $(V = 1)$ of N_2 is small. Experimental results have shown that there is a strong quenching of the $V = 1$ level of N_2 resulting from the (001) level of N_2O.[1] Hence, the pumping reaction given by Eq. (3) is extremely fast, and we assume that these two levels are in local equilibrium. However, this assumption is valid only in the absence of power extraction and can be used for the calculation of small-signal gain where radiative effects are negligible.

In light of the preceding observations, we propose a simplified model for vibrational kinetic processes for the N_2O-N_2

system, as shown in Fig. 1. Here, modes ν_1 and ν_2 are grouped together as mode I and, similarly, mode ν_3 of N_2O and the $V = 1$ level of N_2 are grouped together as mode II. These modes are assumed to be in equilibrium within themselves but not with each other or with the translational mode. Mode I relaxes via τ_c, and mode II relaxes via τ_a and τ_b, where τ_a, τ_b, and τ_c are the average relaxation times of N_2O-N_2O, N_2O-N_2, N_2O-He, N_2-N_2, and N_2-He collisions.

Let $(e_{\text{vib}})_I$ and $(e_{\text{vib}})_{II}$ be the vibrational energies of modes I and II, respectively, per unit mass of the mixture. Then,

$$(e_{\text{vib}})_I = C_{N_2O} R_{N_2O} \left[\frac{\theta_1}{\exp(\theta_1/T_I) - 1} + \frac{2\theta_2}{\exp(\theta_2/T_I) - 1} \right] \qquad (6)$$

$$(e_{\text{vib}})_{II} = C_{N_2O} R_{N_2O} \left[\frac{\theta_3}{\exp(\theta_3/T_{II}) - 1} \right] + C_{N_2} R_{N_2} \left[\frac{\theta_N}{\exp(\theta_N/T_{II}) - 1} \right] \qquad (7)$$

Governing Equations

In the present problem, we have an unsteady, vibrational nonequilibrium, supersonic expansion of a mixture of N_2O, N_2, and He through a convergent-divergent contour nozzle in which the finite-rate molecular energy transfer processes can result in a population inversion between the $(00°1)$ and $(10°0)$ vibrational energy levels of N_2O. We assume the flow to be inviscid and quasi-one-dimensional. The unsteady basic conservation equations governing the flow are

Continuity:

$$\frac{\partial \rho}{\partial t} = -\frac{1}{A} \frac{\partial(\rho u A)}{\partial x} \qquad (8)$$

Momentum:

$$\frac{\partial u}{\partial t} = -\left[\frac{1}{\rho} \frac{\partial p}{\partial x} + u \frac{\partial u}{\partial x} \right] \qquad (9)$$

Energy:

$$\frac{\partial e}{\partial t} = -\left[\frac{p}{\rho} \left(\frac{\partial u}{\partial x} + u \frac{\partial \ln A}{\partial x} \right) + u \frac{\partial e}{\partial x} \right] \qquad (10)$$

State:

$$p = \rho R T \qquad (11)$$

In addition, the rate equations representing the relaxation of vibrational energies of modes I and II are

$$\frac{\partial(e_{\text{vib}})_j}{\partial t} = \frac{1}{\tau_j} [(e_{\text{vib}})_j^{\text{eqb}} - (e_{\text{vib}})_j] - u \frac{\partial(e_{\text{vib}})_j}{\partial x}, \qquad j = \text{I, II} \quad (12)$$

where the superscript eqb indicates the equilibrium vibrational energy that would be contained in mode j at translational temperature.

In Eqs. (12), the relaxation time τ_j characterizes the net rate of energy transfer into and out of mode j. These relaxation times for modes I and II are obtained from

$$\tau_I = \tau_c \qquad (13)$$

$$\frac{1}{\tau_{II}} = \left[\frac{X_{N_2O}}{\tau_a} + \frac{X_{N_2}}{\tau_b} \right] \frac{1}{(X_{N_2O} + X_{N_2})} \qquad (14)$$
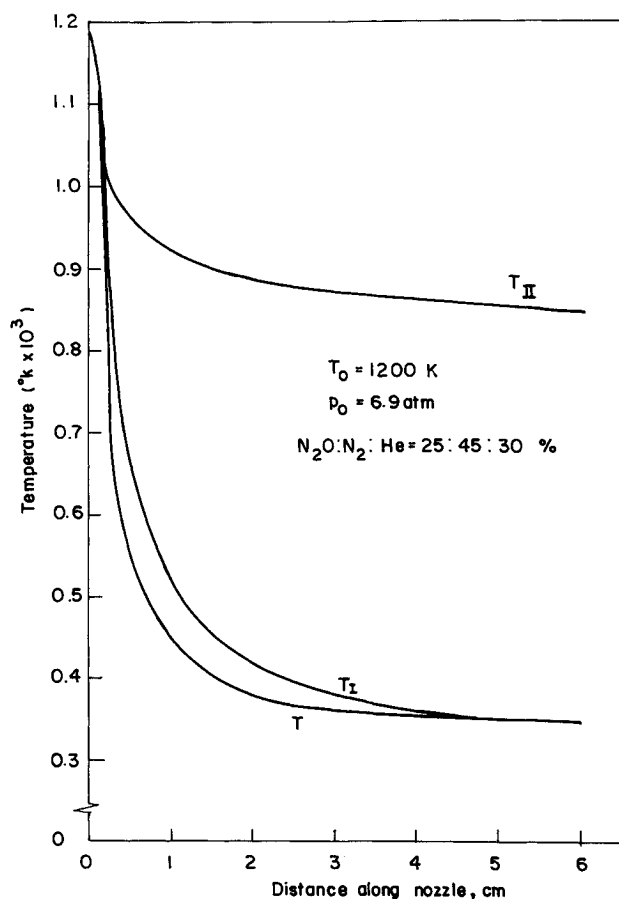
Fig. 2 Steady-state distribution of vibrational and translational temperatures along the nozzle.
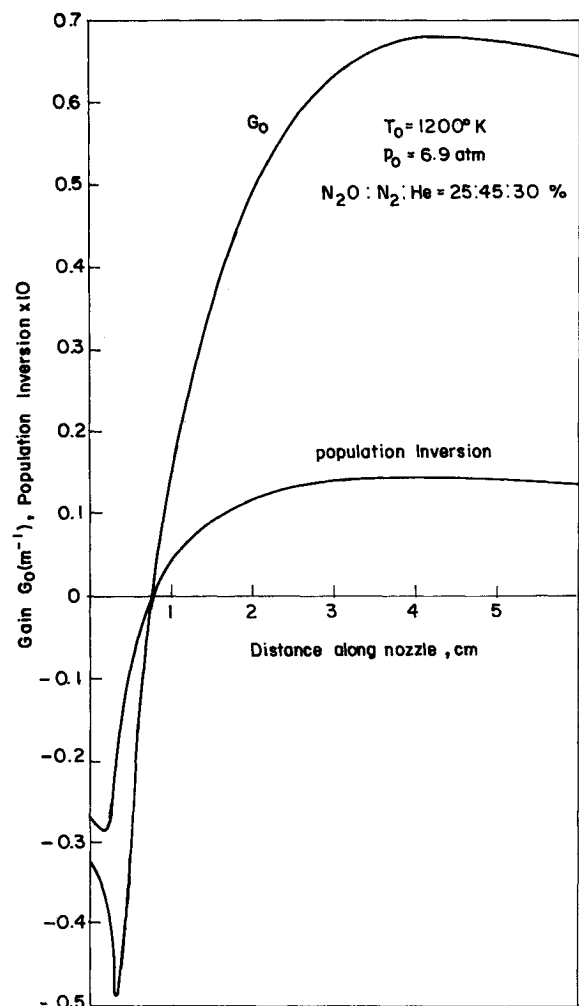

Fig. 3 Steady-state distribution of population inversion and small-signal gain along the nozzle.

where the relaxation times τ_a, τ_b, and τ_c are themselves defined as

$$\frac{1}{\tau_a} = \frac{X_{N_2O}}{(\tau_a)_{N_2O\text{-}N_2O}} + \frac{X_{N_2}}{(\tau_a)_{N_2O\text{-}N_2}} + \frac{X_{He}}{(\tau_a)_{N_2O\text{-}He}} \quad (15)$$

$$\frac{1}{\tau_b} = \frac{X_{N_2O}}{(\tau_b)_{N_2\text{-}N_2O}} + \frac{X_{N_2}}{(\tau_b)_{N_2\text{-}N_2}} + \frac{X_{He}}{(\tau_b)_{N_2\text{-}He}} \quad (16)$$

$$\frac{1}{\tau_c} = \frac{X_{N_2O}}{(\tau_c)_{N_2O\text{-}N_2O}} + \frac{X_{N_2}}{(\tau_c)_{N_2O\text{-}N_2}} + \frac{X_{He}}{(\tau_c)_{N_2O\text{-}He}} \quad (17)$$

In Eqs. (15–17), $(\tau_\beta)_{ki}$, where $\beta = a, b, c, k = N_2O$ for $\beta = a, c$, and $k = N_2$ for $\beta = b$, and $i = N_2O, N_2$, He are the relaxation times of $N_2O\text{-}N_2O$, $N_2O\text{-}N_2$, $N_2O\text{-}He$, $N_2\text{-}N_2$, and $N_2\text{-}He$ collisions. These relaxation times for various collision partners are given in the Appendix for the $N_2O\text{-}N_2\text{-}He$ system.

The vibrational temperatures T_I and T_{II} at steady state are used to compute populations of energy levels within modes I and II, assuming a Boltzmann distribution locally within each mode. Then, these populations are used to compute the small-signal optical gain for the $P(19)$ $(00°1) \rightarrow (10°0)$ transition of the N_2O molecule, using the relation

$$G_0 = \frac{\lambda^2}{4\pi\tau_{12}Z}\left(\frac{23.59}{T}\right)(N_{001} - N_{100})\exp\left(\frac{-229.82}{T}\right) \quad (18)$$

where the collision frequency

$$Z = \sum_i N_i \sigma_{N_2O\text{-}i}\left[\left(\frac{8RT}{\pi}\right)\left(\frac{M_{N_2O} + M_i}{M_{N_2O}M_i}\right)\right]^{1/2} \quad (19)$$

in which the summation is over the chemical species in the laser mixture.

The following data are obtained from the experimental measurements of Ref. 8:

$$\tau_{12} = 3.6 \text{ s}$$

$$\sigma_{N_2O\text{-}N_2O} = 7 \times 10^{-15} \text{ cm}^2$$

$$\sigma_{N_2O\text{-}N_2O} = 7.7 \times 10^{-15} \text{ cm}^2$$

$$\sigma_{N_2O\text{-}He} = 3.6 \times 10^{-15} \text{ cm}^2$$

Numerical Results and Discussions

The governing equations are solved numerically using the time-dependent technique given in Ref. 7. The contour nozzle used in this analysis is described in Ref. 9. The length of the subsonic section is 0.229 cm and that of the supersonic section is 5.744 cm. The area ratios at the inlet and at the exit are 4.2 and 19.75, respectively. In the present studies, 21 grid points consisting of 15 fine and 5 coarse grids were employed through the nozzle. Starting from the initial distribution, the program takes a large number of time steps to approach the final steady-state nonequilibrium results. In this paper, we discuss only the steady-state results.

The results for steady flow of a mixture of N_2O, N_2, and He in a convergent-divergent supersonic contour nozzle are presented in Fig. 2. These curves represent the distribution of translational temperature T, and vibrational temperatures T_I and T_{II} of modes I and II, respectively, along the nozzle at steady state. The curves clearly indicate that mode II relaxes
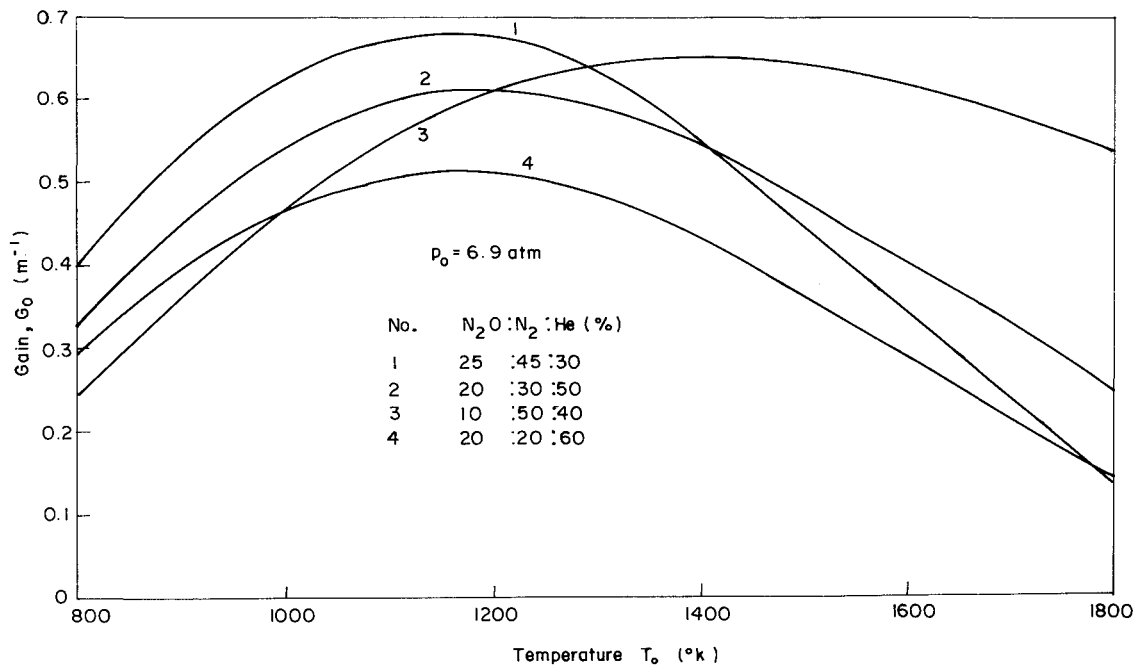
Fig. 4 Variation of small-signal gain at the exit of the nozzle with the plenum temperature for various gas mixture compositions.
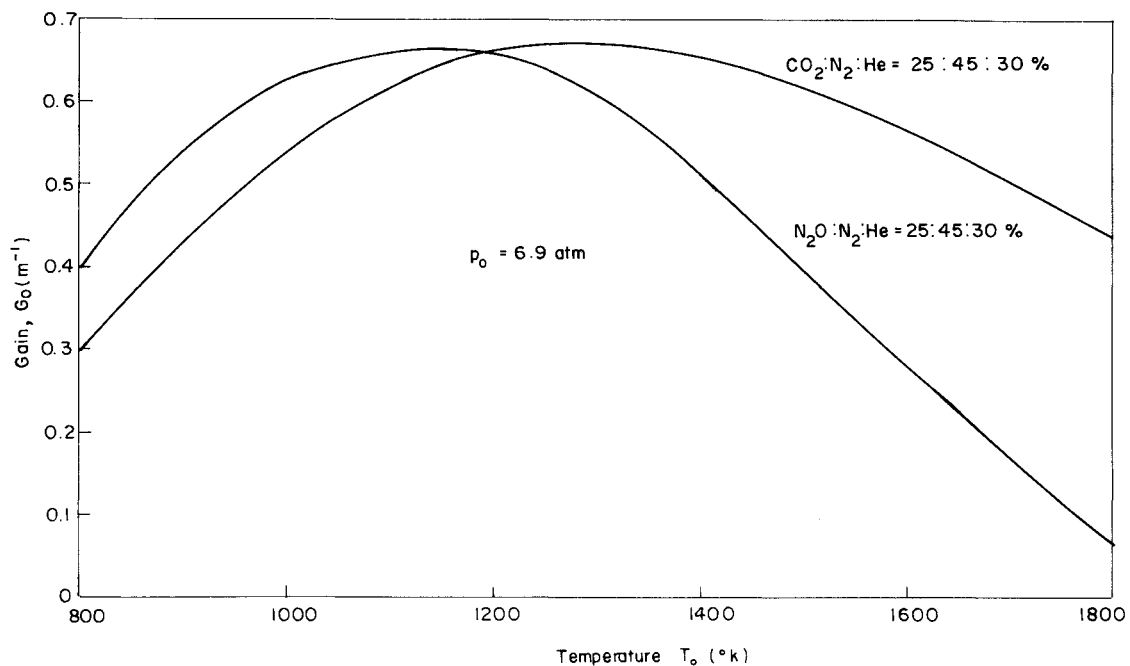

Fig. 5 Comparison of small-signal gain values of N_2O-N_2-He GDL with CO_2-N_2-He GDL.

much more slowly than mode I and that neither of these modes is in equilibrium with the translational temperature.

The temperature distributions shown in Fig. 2 are used, in turn, to compute populations of energy levels within modes I and II, assuming a Boltzmann distribution within each mode. Thus, the population inversion defined as $(N_{001} - N_{100})/N_{N_2O}$ is computed along the nozzle, which is further used to compute the steady-state distributions of small-signal gain G_0 along the nozzle. The distribution of population inversion and G_0 are given in Fig. 3. Downstream of the nozzle throat, the gain decreases rapidly because the spectral line width becomes narrow as a result of the expansion. Thereafter, the gain increases gradually because of the de-excitation of the lower vibrational mode. At about 0.52 cm downstream of the throat, the populations of the (00°1) and (10°0) levels become equal, and hence the population inversion and G_0 are zero. A substantial population inversion develops further downstream

because of the freezing of the higher vibrational mode and the de-excitation of the lower vibrational mode. Thus, the gain increases downstream of the nozzle throat and reaches a maximum value when the lower mode temperature T_I equilibrates with the translational temperature T.

The effect of plenum temperature T_0 on the small-signal gain is investigated, and the results are presented in Fig. 4 for various gas mixture compositions. It is seen that the gain increases with the increase of plenum temperature up to about 1200 K, beyond which it starts decreasing with the further increase of T_0. The highest value of 0.675-m^{-1} small-signal gain is obtained for the gas mixture composition of N_2O:N_2:He = 25:45:30(%), as shown in Fig. 4. The gains of the N_2O GDL and a CO_2 GDL are computed for identical conditions, and the results are presented in Fig. 5. These results clearly indicate that, at lower temperatures, the gain that could be achieved in the N_2O GDL is higher than that in

Table 1 Comparison of the results of time-dependent analysis with the steady-state analysis (Ref. 4)

	Composition:	$X_{N_2O}:X_{N_2}:X_{He} = 0.1:0.4:0.5$
	Initial conditions:	$T_{cr} = 1000$ K
	(at throat)	$P_{cr} = 30$ atm
	Nozzle shape:	$A(x)/A^* = 1 + 20\ell n(1 + x)$

Steady-state values at nozzle exit	Ref. 4	Present analysis
Translational temperature, K	225	243
Vibrational temperature of mode I, K	230	246
Small-signal gain, cm⁻¹	2.5×10^{-2}	2.13×10^{-2}

the CO_2 GDL under identical operating conditions. A lowering of T_0 permits an increase in the efficiency of the N_2O GDL compared with the CO_2 GDL because all the vibrational levels of the N_2O molecule lie below the corresponding levels of the CO_2 molecule and there is a rapid nonresonance exchange in the N_2O laser between (00°1) and the $(V = 1)$ level of N_2, which leads to an increase in the higher level population. This superiority in efficiency breaks down beyond 1200 K.

Biryukov et al.[4] have reported a steady-state analysis of an N_2O-N_2-He GDL using a three-temperature vibrational model for the N_2O-N_2 system. The equations are solved numerically using the Runge-Kutta technique starting from the nozzle throat. In order to compare the results of the present analysis, we have reproduced the results of Ref. 4; these results are shown in Table 1. The gain expression of Ref. 4 is used for small-signal gain computations so as to include the line shape effects. The results given in Table 1 show that the flow parameters and the small-signal gain values predicted by the present analysis closely match those of Ref. 4.

Conclusions

A time-dependent analysis of the N_2O-N_2-He gasdynamic laser is presented. A simplified two-mode model for vibrational energy transfer between N_2O and N_2 molecules is proposed. Numerical results are obtained for the temperatures at steady-state along the supersonic contour nozzle. These results are used to compute the population inversion and the small-signal gain along the nozzle. The gain is shown to become positive after about 0.52 cm downstream of the nozzle throat.

The effect of plenum temperature on the gain of an N_2O GDL is studied, and it is shown that the efficiency of the laser goes down beyond 1200 K. Comparing the performance of the N_2O GDL with the CO_2 GDL reveals that, at lower temperatures, the N_2O GDL is more efficient than CO_2 GDL.

The analysis can be extended for the N_2O-N_2-H_2O mixtures by replacing the He with the H_2O in all the equations and using the appropriate set of vibrational relaxation times.

Appendix: Vibrational Relaxation Times for the N_2O-N_2-He System

The expression for vibrational relaxation times τ_a, τ_b, and τ_c for various collisional partners taken from Ref. 5 are as follows:

$$(p\tau_a)_{N_2O-N_2O} = 0.0163 + 1301.05(T^{-1/3})^{3.724}$$

$$\log(p\tau_a)_{N_2O-N_2} = -12.239 + 19.28(T^{-1/3})^{0.215}$$

$$\log(p\tau_a)_{N_2O-He} = -12.189 + 19.28(T^{-1/3})^{0.215}$$

$$\log(p\tau_b)_{N_2-N_2} = 93(T^{-1/3}) - 4.61$$

$$\log(p\tau_b)_{N_2-He} = 60.7(T^{-1/3}) - 4.168$$

$$(p\tau_b)_{N_2-N_2O} = (p\tau_b)_{N_2-N_2}$$

$$(p\tau_c)_{N_2O-N_2O} = -0.0999 + 131.38(T^{-1/3})^{2.535}$$

$$(p\tau_c)_{N_2O-N_2} = -0.1689 + 193.537(T^{-1/3})^{2.496}$$

$$(p\tau_c)_{N_2O-He} = 0.0592 + 8.3 \times 10^6(T^{-1/3})^{10.56}$$

In the preceding relations, $(p\tau)$ is in atmospheres per microsecond, and the temperature T is in degrees Kelvin.

References

[1]Patel, C. K. N., "CW Laser Action in N_2O (N_2-N_2O System)," *Applied Physics Letters*, Vol. 6, Jan. 1965, pp. 12–13.

[2]Fox, K. E. and Reid, J., "Dynamics of the N_2O Laser as Measured with a Tunable Diode Laser," *Journal of the Optical Society of America, B*, Vol. 2, May 1985, pp. 807–814.

[3]Milewski, J. et al., "Experiment with a CW N_2O Gasdynamic Laser and Its Qualitative Comparsion with a CO_2 Laser," *Applied Physics Letters*, Vol. 30, April 1977, pp. 342–344.

[4]Biryukov, A. S., Kulagin, Y. A., and Shelepin, L. A., "Kinetics of the N_2O Gasdynamic Laser," *Soviet Physics—Technical Physics*, Vol. 21, Feb. 1976, pp. 199–202.

[5]Masuda, W., Yamada, H., and Yamane, M., "Gain Characteristics of an N_2O Gasdynamic Laser," *Transactions of the Japan Society for Aeronautical and Space Sciences*, Vol. 28, May 1985, pp. 44–53.

[6]Masuda, W., Yamada, H., and Ogawahara, N., "Numerical Simulation of an N_2O Downstream Mixing Gasdynamic Laser," *Bulletin of the Japan Society of Mechanical Engineers*, Vol. 29, Oct. 1986, pp. 3442–3447.

[7]Anderson, J. D., Jr., "Time-Dependent Analysis of Population Inversion in an Expanding Gas," *Physics of Fluids*, Vol. 13, Aug. 1970, pp. 1983–1989.

[8]Djeu, N., Kan, T., and Wolga, G., "G10-Laser Parameters for the 10.8-μ N_2O Molecular Laser," *IEEE Journal of Quantum Electronics*, Vol. QE-4, Nov. 1968, pp. 783–785.

[9]Glowacki, W. J. and Anderson, J. R., Jr., "A Computer Program for CO_2-N_2-H_2O Gasdynamic Laser Gain and Maximum Available Power," Naval Ordnance Laboratory, White Oak, MD, NOLTR 71-210, 1971.