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PERFORMANCE AND DIAGNOSTIC MEASUREMENTS OF A SUPERSONIC HCL CHE--ETC(U)
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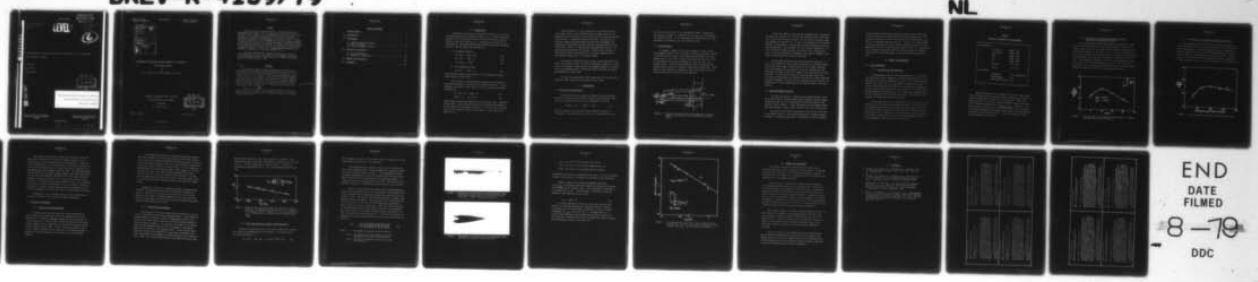

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PERFORMANCE AND DIAGNOSTIC MEASUREMENTS OF A SUPERSONIC HCl CHEMICAL LASER

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PERFORMANCE AND DIAGNOSTIC MEASUREMENTS OF A SUPERSONIC HCl CHEMICAL LASER

by

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RESUME

On décrit un laser chimique au HCl, à régime continu et à écoulement supersonique, dont la concentration d'atomes de chlore en déséquilibre est produite par l'interaction du NO avec du ClO_2. Le ClO_2 est injecté dans un écoulement de NO dilué dans l'hélium, immédiatement en amont de l'étranglement d'une tuyère à fente. Les gaz en réaction subissent alors une expansion de jet libre dans une région de basse pression où du HI est ajouté pour produire rapidement du HCl excité dans des niveaux de vibration. La puissance de sortie maximum de 35W obtenue dans plusieurs raies du HCl représente une efficacité chimique de quelque 4.5% basée sur le dégagement exothermique de la réaction de pompage et le taux d'écoulement du HI.

On décrit également une technique efficace pour visualiser l'écoulement dans la zone d'expansion. Cette technique est basée sur la chimiluminescence visible résultant de l'addition de H_2S au système de réaction NO/ClO_2. (NC)

ABSTRACT

A supersonic purely chemical continuous wave HCl laser is described in which nonequilibrium concentrations of atomic chlorine are generated by the branch-chain reaction of NO with ClO_2. The ClO_2 is injected into a stream of NO in helium immediately upstream of the throat of a slit nozzle. The reacting gases then expand as a free-jet into a low-pressure region where HI is added to rapidly form vibrationally excited HCl. The maximum HCl multiline output power of 35 W obtained represents a chemical efficiency of ~ 4.5% based on the total exothermicity of the pumping reaction and the limiting flow rate of HI.

An efficient technique for visualizing the flow in the expanded region is described. This technique is based on the visible chemiluminescence resulting from the addition of H_2S to the NO/ClO_2 reaction system. (U)

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1.0 INTRODUCTION

The operation of a purely chemical HCl laser at 3.6-4.0 μm was recently reported by this laboratory (Refs. 1,2). Chlorine atoms were produced by the branch-chain reaction of nitric oxide with chlorine dioxide in the molar ratio of 2:1. Vibrationally excited HCl, the lasing medium, was formed when hydrogen iodide reacted with the atomic chlorine. The principal reaction steps are:

a) Prepumping chemistry

$$NO + ClO_2 \rightarrow NO_2 + ClO \quad [1]$$

$$NO + ClO \rightarrow NO_2 + Cl \quad [2]$$

$$Cl + ClO_2 \rightarrow 2\ ClO \quad [3]$$

b) Laser pumping reaction

$$Cl + HI \rightarrow HCl^{\dagger} + I \quad [4]$$

A detailed description of the kinetics of the $NO/ClO_2/HI$ chemical laser system is given in Ref. 2.

The efficiency of the prepumping chemistry may be reduced if the reaction sequence is carried out at elevated pressures. The most important loss process for Cl atoms is the NO_2 catalysed three-body recombination:

$$Cl + NO_2 + M \rightarrow ClNO_2 + M \quad [5]$$

$$ClNO_2 + Cl \rightarrow Cl_2 + NO_2 \quad [6]$$

which becomes increasingly important as the pressure rises. Modelling studies (Ref. 2) have shown that the rate of chlorine atom formation by the branch-chain mechanism, reactions 1-3, and the rate of chlorine atom loss by reactions 5 and 6 become equal at a total pressure of about 13 torr.

Self-relaxation is a limiting process in HCl chemical lasers. In CW laser devices, self-relaxation places an upper limit on the population inversion density of HCl, that can be sustained for a given flow velocity. A transverse flow HCl laser may, in principle, be scaled to higher powers (and higher resultant mass throughputs) by widening the flow channel and increasing the pumping capacity of the flow system. However, to maintain the same approximate device size, increased flow velocities are required. Usually, this increase in flow velocity entails acceleration of the gas by expansion from a high pressure plenum into a low pressure region.

For a purely chemical HCl laser system, these requirements may be met by designing a laser in which the prepumping chemistry is initiated in the subsonic region and the laser pumping chemistry is carried out in the expanded region. The present report describes the design and operation of such a laser.

This work was performed at DREV between May 1977 and June 1978 under PCN 33H07, Research on Chemically Excited Lasers.

2.0 EXPERIMENTAL

2.1 Chlorine dioxide generator

Chlorine dioxide was prepared on demand by passing molecular chlorine in diluent helium through a column containing $NaClO_2$ flakes. The Cl_2 was converted to ClO_2 via the heterogeneous reaction

$$2\ NaClO_2 + Cl_2 \rightarrow 2\ NaCl + 2\ ClO_2\ . \qquad [7]$$

The ClO_2 effluent could enter the laser apparatus in either of 2 locations. Further diluted with helium, it constitutes the main

core flow to which NO and HI are subsequently added. Alternately, the ClO_2 may be added as a secondary gas to a main core flow consisting of NO and helium. In the experiments described herein, the ClO_2 was added as a secondary gas unless otherwise noted.

2.2 Nozzle assembly

A schematic diagram of the nozzle assembly is shown in Fig. 1. The prepumping chemistry was initiated by injecting ClO_2 into a stream of NO and helium at a point 1.5 mm upstream of the throat of a converging nozzle. The cross section of the throat was 25 cm by 1.2 mm. The reacting NO and ClO_2 then expanded as a free-jet into a low pressure region. After allowing sufficient time for chlorine atom formation HI was injected to produce vibrationally excited HCl. Laser power was then extracted by defining a laser cavity whose axis was slightly downstream of the point of HI addition.

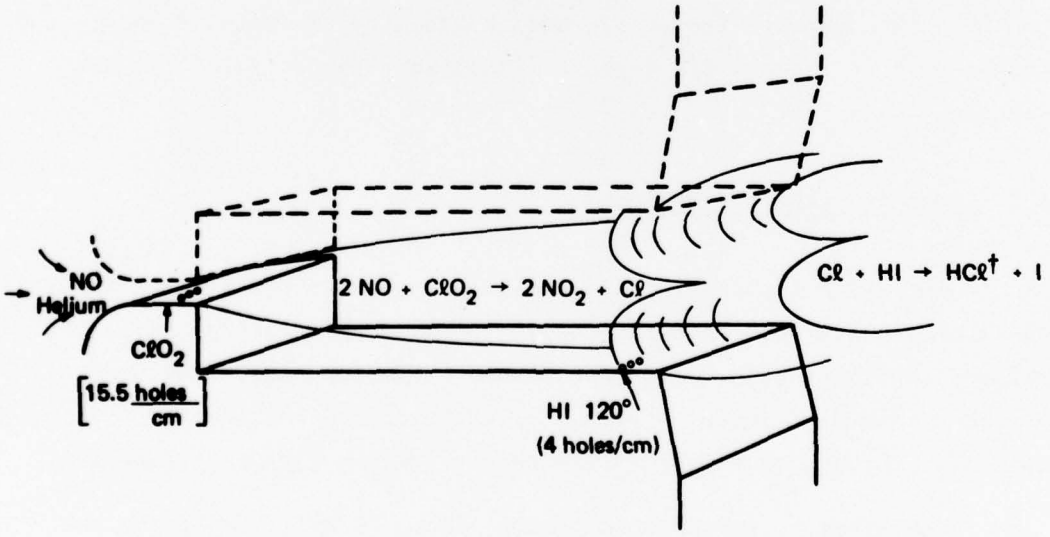

FIGURE 1 - Schematic diagram of the nozzle assembly for a purely chemical HCl laser based on the $NO/ClO_2/HI$ reaction system

In both the subsonic region and the expanded region, the secondary gases were injected into the main flow stream as minute individual jets issuing from the wall structure. At the subsonic location, where the pressure was typically 25 - 35 torr, the secondary gas was injected through 776 hemispherical orifices equally spaced along the 25-cm-long nozzle (388/side). Each hemispherical orifice had an opening of 2.5 x 10^{-2} cm^2 and was formed by mating a flat plate with a multigrooved plate. With this demountable arrangement, the injector assembly could easily be taken apart and cleaned if necessary.

In the expanded region, where the pressure was typically 2 - 3 torr, the secondary gas (HI diluted with helium) was injected through 198 injector holes equally spaced along the 25-cm length (99/side). Each 1-mm-diameter hole was oriented so that the secondary gas was injected at an angle of 120^{o} with respect to the direction of the main core flow. It was believed that this injection angle would allow increased jet penetration into the main core flow for a given secondary gas mass throughput. The entire nozzle assembly was made of monel, and formed part of a monel flange that fitted into the entry of the laser section.

2.3 Laser and exhaust sections

The laser section was a stainless steel housing having a 15-by 45-cm cross-section. Brewster angle windows mounted along the length allowed the laser axis to be established at various distances downstream of the HI injector. Visual observations of the expanded region were made through an acrylic plate located on the top of the housing.

The overall cross-section increased to 30 by 45 cm at the entry to the exhaust section. This section consisted of a finned precooler followed by a liquid nitrogen trap. The precooler served to minimize

liquid nitrogen consumption by removing as much of the reaction heat as possible before the gases reached the liquid nitrogen trap. The precooler consisted of a series of 25 monel fins stacked side by side with a 1 cm separation between each fin. In place, the fins extended 5 cm above and below the center plane as defined by the nozzle. The dimension of the fins in the main flow direction was 8 cm. Receptacles for 4 commercial heat pipes, each of which was nominally capable of removing ∿300 W of thermal power, were attached above and below the fins.

3.0 RESULTS AND DISCUSSION

3.1 Laser performance

3.1.1 Conditions for maximum power

The maximum multiline HCl laser output power of 35 W obtained from the present device represents a chemical efficiency of ∿4.5 percent based on the total exothermicity of the Cl + HI reaction and the limiting flow rate of HI. The experimental conditions are given in Table I where helium (flow 1) refers to the helium that accompanied the NO to form the main core flow, and helium (flow 2) refers to the helium that was mixed with the HI. The secondary gas (helium (flow 2)/HI) was injected at 120^{o} to the main core flow. The laser axis was located 1 cm downstream from the HI injector.

The optimum flow of HI was determined for a given NO/ClO_2 mix by adding HI until the laser power stopped increasing. This flow of HI was slightly lower than the stoichiometric amount required to convert all of the Cl present into HCl, so that the HI flow rate became the limiting flow rate. Conversion of less than 100% of the Cl_2 into ClO_2 in the $NaClO_2$ column and/or a failure of the prepumping reaction to reach completion, could explain this observation.

TABLE I

Supersonic chemical laser performance

Flow Conditions			
	He (flow 1)	135000	sccm
	He (flow 2)	14000	sccm
	$C\ell O_2$	11000	sccm
	NO	22000	sccm
	HI	8400	sccm
	P_{plenum}	32	torr
	P_{cavity}	2.7	torr
Laser Output			
	Decoupling	15%	transmission
	Maximum power	35 W	
	Chemical efficiency	4.5%	

Optimum performance was obtained for relatively large helium flows indicating that a considerable amount of helium was required to moderate temperature increases due to the reaction. This is not surprising since fluid dynamic cooling should be slight owing to the modest expansion pressure ratio, P_{plenum}/P_{cavity} of about 10. The balance between helium (flow 1) and helium (flow 2) was dictated by the optimum momentum balance between the secondary gas stream and the mainstream (Cl,NO_2/helium (flow 1)). The rate of mixing of these streams depends on this balance.

3.1.2 Laser power as a function of distance between the laser axis and the HI injector

Total HCl multiline laser output power was measured as a function of the distance between the laser axis and the HI injector, Z. Figure 2 shows a plot of laser power versus distance, obtained with an 8-mm defining aperture in the laser cavity. The power increases at a rate that is probably determined by the rate of mixing of the secondary stream (helium (flow 2)/HI) with the mainstream (helium (flow 1)/Cl, NO_2). After reaching a maximum at Z=1 cm, the power decreases until it is about half of the maximum power at Z=2 cm. This fall-off is believed to be due to rapid self-relaxation processes of vibrationally excited HCl which tend to destroy the population inversion.

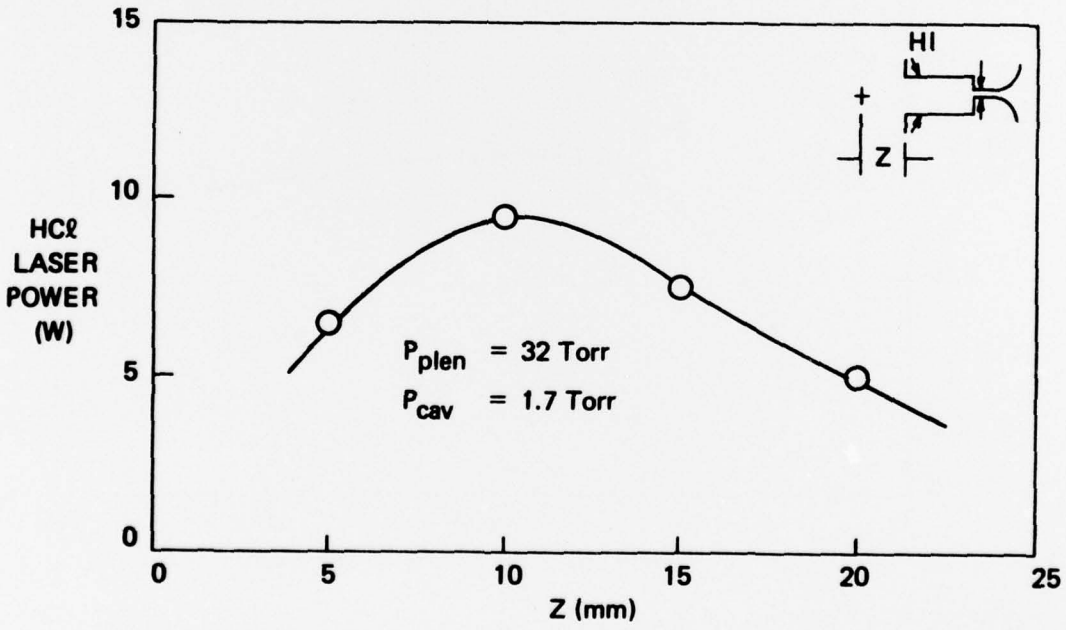

FIGURE 2 - HCl laser power as a function of the distance, Z, between the laser axis and the HI injector

3.1.3 Laser power as a function of plenum helium flow

The dependence of total HCl multiline laser output power on plenum helium flow (helium (flow 1)) was studied in an earlier geometric configuration. In this configuration, the HI injector was located 7 mm from the throat and the HI was injected at an angle of 90^{o} with respect to the direction of the main core flow. Figure 3 shows a plot of laser power versus plenum helium flow. Apparently, a certain amount of helium is required, probably for cooling the laser

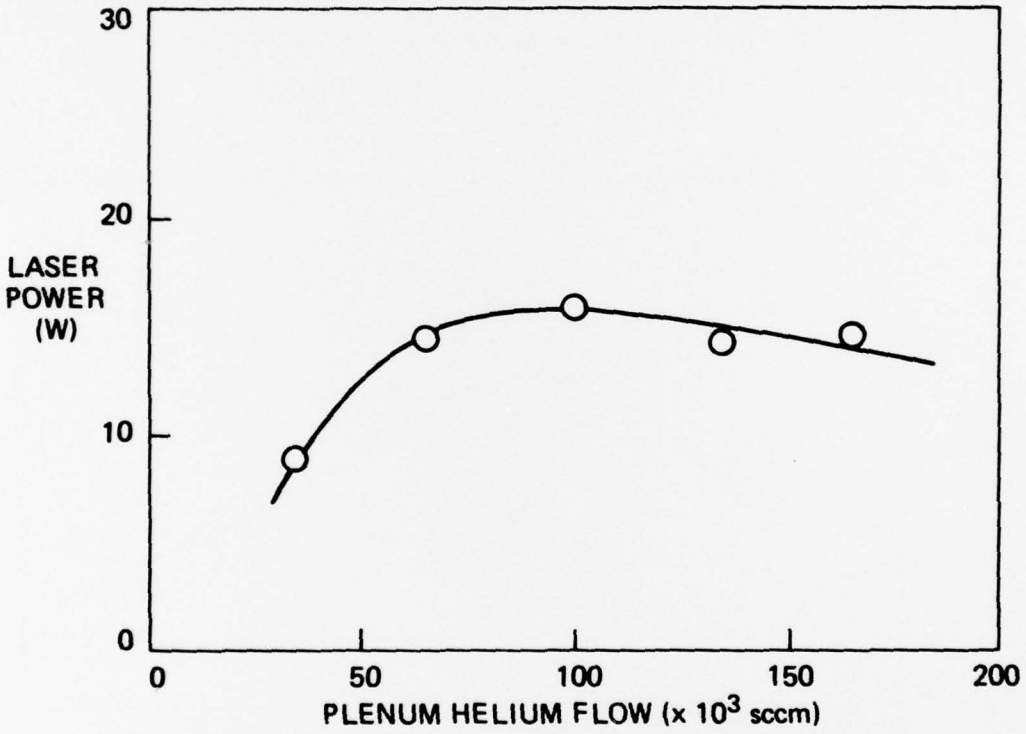

FIGURE 3 - HCl laser power as a function of plenum helium flow

medium as evidenced by the rising portion of the curve. The fact that the laser power falls off only marginally at helium flows up to 2×10^5 sccm, which corresponds to a plenum pressure of ~50 torr, is somewhat surprising since the loss of chlorine atoms by three-body recombination should be quite severe at this pressure. One explanation for this observation is that most of the chlorine atoms are produced after passage through the throat and during expansion where the pressure is much lower. Evidence supporting this explanation is given in the next Section.

3.1.4 HCl laser output spectra: saturation effects

The multiline HCl laser output was analyzed by sampling a portion of the output beam with a MacPherson model 218 0.3-mm monochromator, equipped with a 150 lines/mm grating blazed at 3 μm. The signal was chopped at 670 Hz, detected with an InSb detector and amplified with a Princeton Applied Research (HR-8) lock-in amplifier. All measurements were made with the laser axis located 1 cm from the HI injector.

Spectra were taken at 3 different decoupling fractions. Nominal mirror transmissions were 2%, 10% and 20%. The total output powers for these 3 mirrors were 2.5, 7.5 and 6.0 W respectively. Laser power originated from P branch lines of $H^{35}Cl$ from the $v=3 \rightarrow 2$, $v=2 \rightarrow 1$ and $v=1 \rightarrow 0$ bands and from the $v=2 \rightarrow 1$ and $v=1 \rightarrow 0$ bands of $H^{37}Cl$. The observed lines together with their relative approximate contributions to the total power are given in Table II.

TABLE II

Supersonic HCℓ laser spectral output

Transmission λ (μm)		∿ 2% Intensity		∿ 10% Intensity		∿ 20% Intensity	
		$H^{35}Cℓ$	$H^{37}Cℓ$	$H^{35}Cℓ$	$H^{37}Cℓ$	$H^{35}Cℓ$	$H^{37}Cℓ$
3.9538	$P_{3-2}(7)$	2.0	--	4.2	--	2.8	--
3.9181	(6)	25.8	--	22.2	1.5	18.2	--
3.8839	(5)	37.5	6.5	27.8	2.8	33.0	--
3.8511	(4)	20.5	2.5	1.5	--	10.0	--
3.8050	$P_{2-1}(7)$	13.5	--	15.5	--	16.3	--
3.7711	(6)	32.8	3.0	46.8	2.0	51.6	--
3.7385	(5)	50.8	8.2	48.2	5.5	59.2	--
3.7072	(4)	12.5	7.2	14.2	6.5	40.7	--
3.6660	$P_{1-0}(7)$	18.5	--	9.5	--	--	--
3.6337	(6)	37.5	--	31.0	--	29.0	--
3.6026	(5)	20.0	--	38.8	--	65.6	--
3.5728	(4)	14.5	--	7.0	--	14.3	--
% $v = 3 \rightarrow 2$		30.3		21.0		18.8	
$v = 2 \rightarrow 1$		40.9		48.7		49.3	
$v = 1 \rightarrow 0$		28.9		30.3		32.0	

The relative contribution of each of the separate vibrational bands tends to equalize with decreasing output coupling. This is believed to be due to the saturation of the medium and to a decrease in the threshold gain. With low saturation (20% output coupling), the v=2→1 lines contribute most to the total laser power suggesting that the highest gain lines originate from this band. As the medium saturation increases with 10% and 2% output coupling, the relative contribution of v=2→1 lines decreases and that of the v=3→2 lines increases. Evidently, strong lasing of the v=2→1 transition decreases the population of v=2 thereby increasing the inversion of the v=3→2 transitions. The fraction of total HCl laser power in the v=1→0 band is relatively insensitive to output coupling, implying that the population of both v=1 and v=0 changes in a similar way with increasing medium saturation.

Consequently, with a suitable choice of mirror transmission, the power of specific laser transitions could be maximized.

3.2 Diagnostic experiments

3.2.1 Plenum pressure measurements

The observation that laser power was largely unaffected by plenum pressure up to pressures of ∿50 torr suggested that in the present laser, the NO/ClO_2 prepumping chemistry had not proceeded to a great degree by the time the gases reached the throat of the nozzle. Evidence supporting this suggestion was obtained by indirectly measuring the reaction heat evolved. If heat is evolved upstream of the throat, 'thermal blockage' should occur due to the sudden increase in enthalpy of the gases leading to an increase in the total plenum pressure.

An experiment was performed in which plenum pressure measurements were compared a) for the case where no reaction had occurred and b) for the case where the NO/ClO_2 reaction was allowed to proceed. In this experiment, blockage effects of a purely physical nature were taken into account by using, in place of ClO_2, an 'inert' dummy gas of approximately the same molecular weight as ClO_2, namely Cl_2, for the 'no reaction' case. For complete reaction between NO and ClO_2, a pressure increase of ∿9.3 torr would have been observed. The fact that the measured plenum pressures for the 2 cases were almost identical indicates that little 'thermal blockage' occurred in the throat region.

Apparently, most of the prepumping chemistry transpired after passage of the gases through the throat and during the expansion process. The line of ClO_2 injection orifices was so close to the throat (1.5 mm) that at an ambient pressure of ∿25 torr, mixing did not occur fast enough to allow extensive reaction between ClO_2 and NO before the gases reached the throat.

3.2.2 Burn pattern measurements

Burn patterns of the laser output beam were obtained by placing a lucite block directly behind the decoupling mirror. Since these burn patterns generally tended to reflect the approximate spatial distribution of the active medium, they were utilized to obtain information about the free-jet geometry. In a free-jet, the width of the issuing main core flow depends on the ratio of the pressure in the plenum region to the pressure in the expanded region. Since the gain region should be limited to the breadth of the core flow, the width of the laser burn pattern may be expected to depend similarly on this pressure ratio. Thus, for a given plenum pressure, the width of the laser burn

pattern should decrease as the cavity pressure is increased. This expected narrowing of the laser burn pattern was observed, and is shown in Fig. 4. In this experiment, 2 mirrors having a 4-m radius of curvature were separated by 60 cm. The total decoupling fraction was ∿20%.

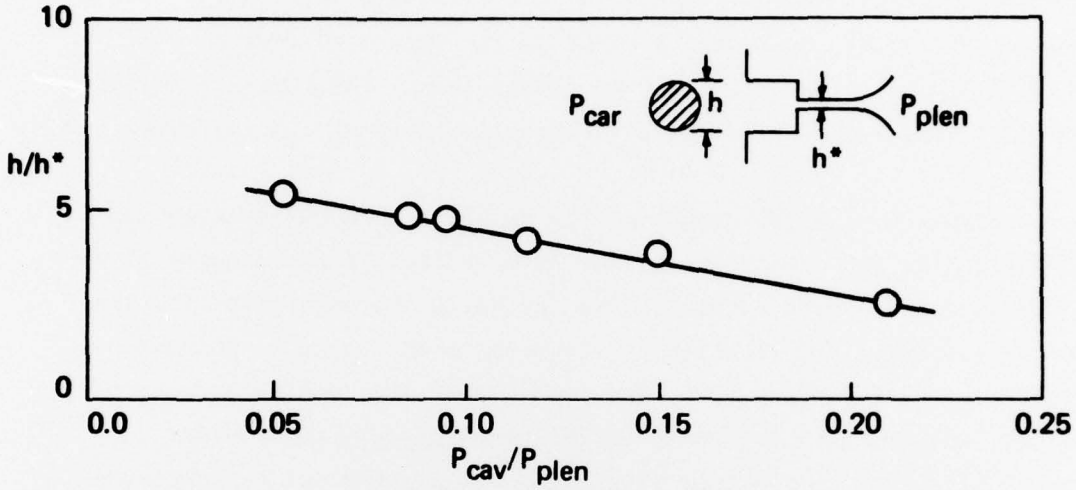

FIGURE 4 - Width of the laser burn pattern as a function of cavity pressure. The width of the burn pattern, h, is expressed as a multiple of throat heights, h*. The cavity pressure, P_{cav}, is expressed as a fraction of the plenum pressure, P_{plen}.

3.2.3 Flow visualization by visible chemiluminescence

When HI was replaced by H_2S, no HCl laser emission was obtained but a blue-red chemiluminescence was observed. The initial step in the reaction sequence is:

$$Cl + H_2S \rightarrow HCl + HS \quad k = 6.0 \times 10^{-11} cm^3 s^{-1} \quad [3] \qquad [8]$$

The subsequent reactions and the emitting species responsible for the chemiluminescence have yet to be identified.

The chemiluminescence, however, can be used to visualize the flow in the expanded region. Mixing phenomena near the H_2S injectors are clearly visible as shown in Fig. 5. This photograph was taken from above (perpendicular to the flow direction and laser axis), and shows a portion of the reacting gases in the expansion region. Individual H_2S injector locations are easily seen. The individual jets tended to coalesce $\sim$2 cm downstream from the point of H_2S injection.

Figure 6 is a photograph of the $NO/ClO_2/H_2S$ chemiluminescence taken from the side (parallel to the laser axis). The secondary flows of H_2S in helium which entered above and below the main core flow are clearly visible. The location of the point where the 2 luminous zones merged was easily determined. The location of this coalescence point was very sensitive to the ratio of the secondary gas (H_2S/helium (flow 2))flow rate to the core flow rate. This ratio may be expressed as the momentum flux ratio (MFR) which is the ratio of momentum per unit area through the individual secondary orifices (subscript s) to that in the main core flow (subscript ∞). If the flow velocities are approximately sonic, the MFR may be written in terms of the molar flow rates as

$$MFR = \frac{1}{N}\left(\frac{F_s}{F_\infty}\right)\left(\frac{A_\infty}{A_s}\right)\left(\frac{\gamma_s}{\gamma_\infty}\right)^{\frac{1}{2}}\left(\frac{T_s}{T_\infty}\right)^{\frac{1}{2}}\left(\frac{M_s}{M_\infty}\right)^{\frac{1}{2}} \qquad [9]$$

where N = the number of secondary injector orifices,

F_s/F_∞ = the ratio of total molar flow rate in the secondary flow to that in the main flow,

A_s/A_∞ = the ratio of secondary orifice area to the core-flow area,

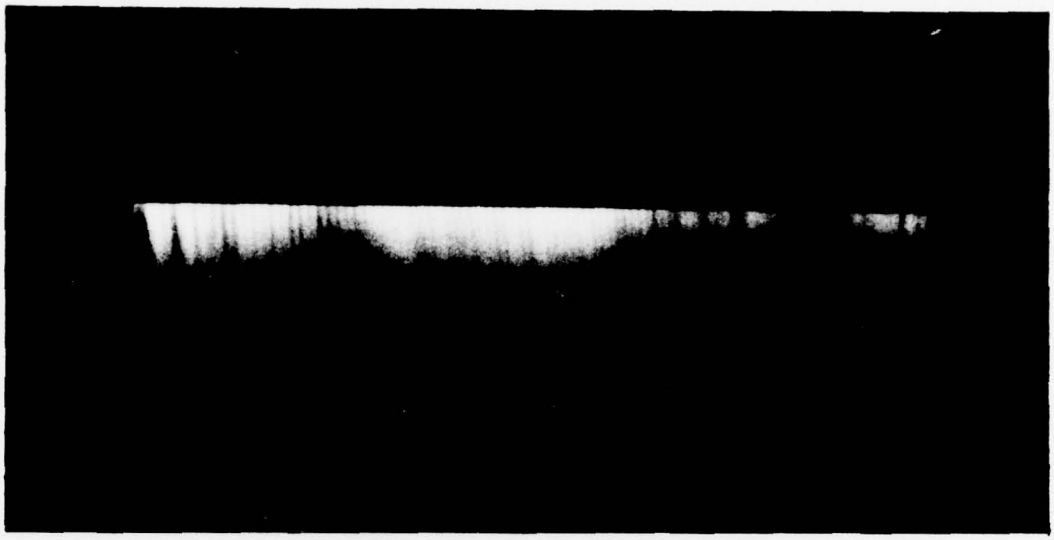

FIGURE 5 - Photograph of the $NO/ClO_2/H_2S$ chemiluminescence taken from above (perpendicular to the flow direction and laser axis). Scale: four H_2S jets per cm

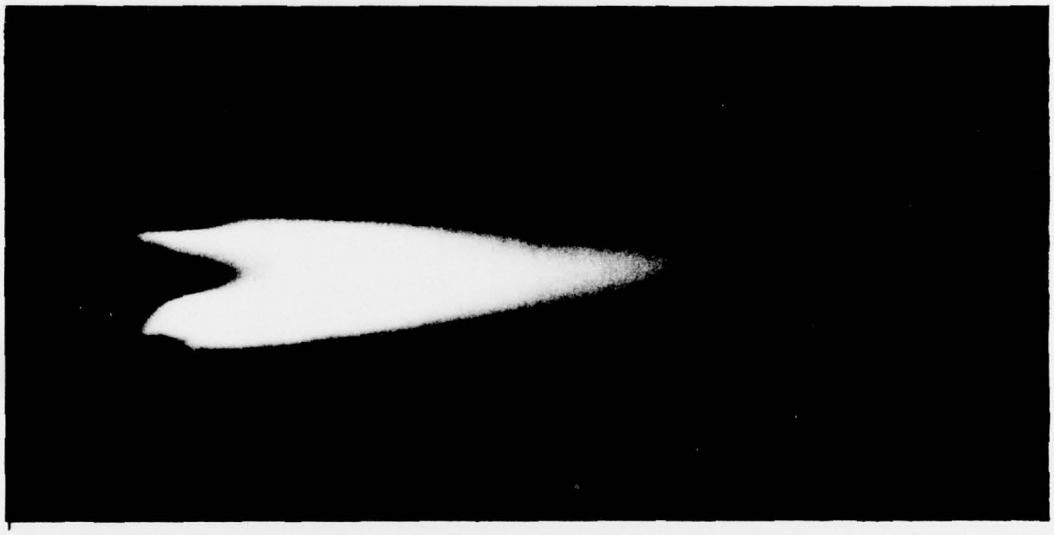

FIGURE 6 - Photograph of the $NO/ClO_2/H_2S$ chemiluminescence taken from the side (parallel to the laser axis)

γ_s/γ_∞ = the ratio of the specific heat ratios,

T_s/T_∞ = the ratio of the absolute temperatures, and

M_s/M_∞ = the ratio of the average molecular weights.

The momentum flux ratio was found previously (Ref. 4) to be a convenient indicator of the extent of secondary jet penetration into main flows.

Figure 7 shows a plot of MFR, the momentum flux ratio, versus Z_{mid}, the distance from the secondary gas injector to the point of confluence of the luminous zones. (For calculations of the MFR, it was assumed that $T_s \sim T_\infty$). This distance is evidently a sensitive function of the momentum flux ratio with

$$Z_{mid} \propto (MFR)^{-1.8} \qquad [10]$$

For laser experiments with HI as a fuel, the laser axis was located 1 cm from the secondary gas (HI/helium (flow 2)) injector. For optimum laser flow conditions, the MFR has a value of $\sim$3 which corresponds to a Z_{mid} $\sim$8 mm (see Fig. 7). Although there is no visible chemiluminescence from the $NO/ClO_2/HI$ reaction system, the zone where the 2 entering HI flows meet is apparently almost coincident with the optimum location of the laser axis.

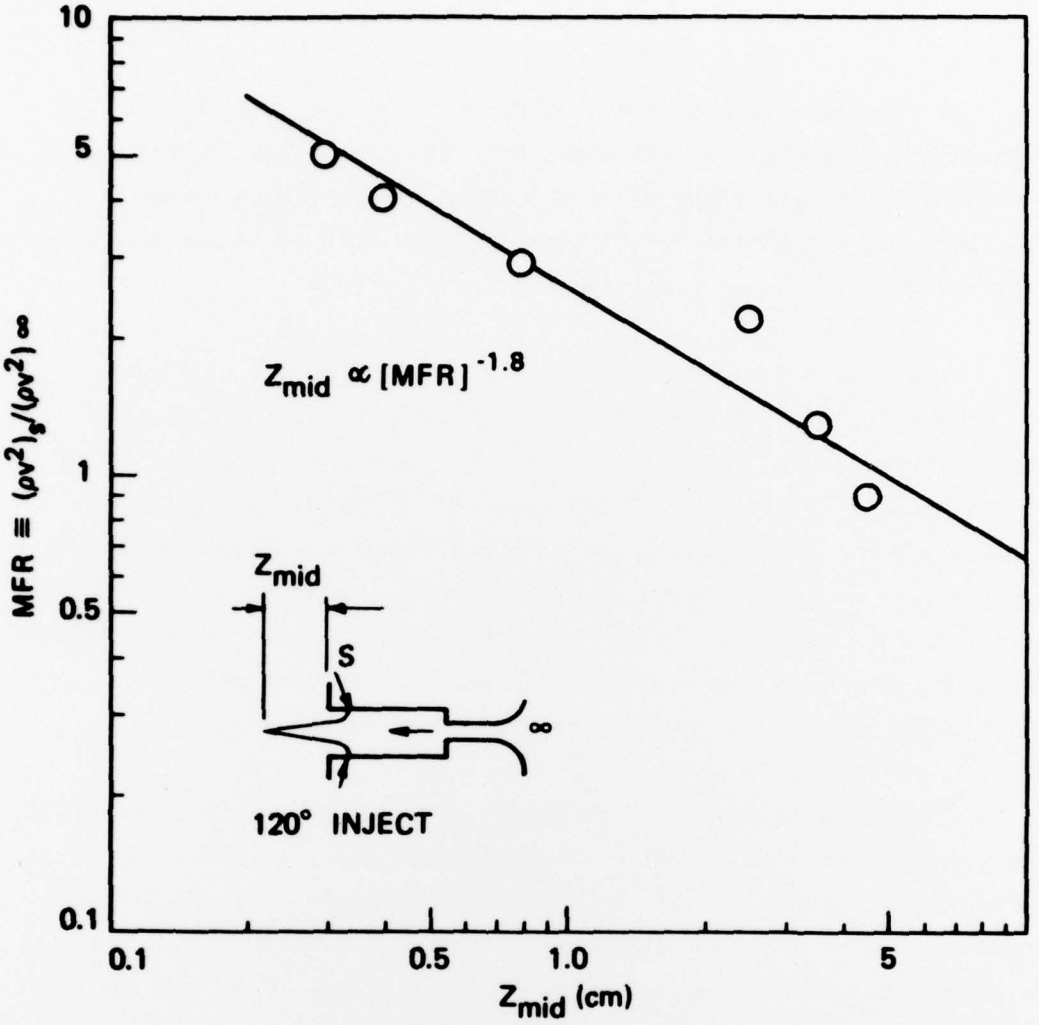

FIGURE 7 - The momentum flux ratio, MFR, versus distance from the secondary gas injector to the point of confluence of the luminous zone, Z_{mid}, for the $NO/ClO_2/H_2S$ reaction

4.0 SUMMARY AND CONCLUSIONS

A supersonic chemical HCl laser based on the reaction of chemically generated chlorine atoms with HI was studied. The maximum HCl multiline output power of 35 W obtained represents a chemical efficiency of $\sim$ 4.5% based on a limiting flow rate of HI and the total exothermicity of the pumping reaction.

This laser device incorporated a simple free-jet expansion. The prepumping NO/ClO_2 chemistry was initiated by wall injection of one of the reagents just upstream of the nozzle throat. Experiments indicated that most of the NO/ClO_2 reaction occurred during the expansion process after passage through the throat. Apparently, mixing was too slow for substantial reaction to occur upstream of the throat. Consequently, successful laser operation was obtained at higher plenum pressures than those expected on the basis of calculations of the loss of chlorine atoms by NO_2-catalysed three-body recombination.

The spectral output of the laser occurred between 3.6 and 4.0 μm and consisted of vibrational-rotational transitions from the v=3→2, v=2→1 and v=1→0 vibrational bands of $H^{35}Cl$ and the v=2→1 and v=1→0 bands of $H^{37}Cl$. A power extraction effect was observed in which the relative contributions of the individual vibrational bands to the total power tended to equalize as the overall medium saturation increased.

The rate of mixing in the expanded region was inferred from flow-visualization measurements using H_2S as a fuel instead of HI. Optimum mixing is believed to depend critically on the momentum flux balance between the main core flow which contains chlorine atoms and the injected secondary gas flow which contained HI.

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DREV, P.O. Box 880, Courcelette, Que. G0A 1R0

"Performance and Diagnostic Measurements of a Supersonic HCl Chemical Laser"
by - S.J. Arnold, K.D. Foster and D.R. Snelling

A supersonic purely chemical continuous wave HCl laser is described in which nonequilibrium concentrations of atomic chlorine are generated by the branch-chain reaction of NO with ClO_2. The ClO_2 is injected into a stream of NO in helium immediately upstream of the throat of a slit nozzle. The reacting gases then expand as a free-jet into a low-pressure region where HI is added to rapidly form vibrationally excited HCl. The maximum HCl multiline output power of 35 W obtained represents a chemical efficiency of $\sim$ 4.5% based on the total exothermicity of the pumping reaction and the limiting flow rate of HI.

An efficient technique for visualizing the flow in the expanded region is described. This technique is based on the visible chemiluminescence resulting from the addition of H_2S to the NO/ClO_2 reaction system. (U)

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On décrit un laser chimique au HCl, à régime continu et à écoulement supersonique, dont la concentration d'atomes de chlore en déséquilibre est produite par l'interaction du NO avec du ClO_2. Le ClO_2 est injecté dans un écoulement de NO dilué dans l'hélium, immédiatement en amont de l'étranglement d'une tuyère à fente. Les gaz en réaction subissent alors une expansion de jet libre dans une région de basse pression où du HI est ajouté pour produire rapidement du HCl excité dans des niveaux de vibration. La puissance de sortie maximum de 35W obtenue dans plusieurs raies du HCl représente une efficacité chimique de quelque 4.5% basée sur le dégagement exothermique de la réaction de pompage et le taux d'écoulement du HI.

On décrit également une technique efficace pour visualiser l'écoulement dans la zone d'expansion. Cette technique est basée sur la chimiluminescence visible résultant de l'addition de H_2S au système de réaction NO/ClO_2. (NC)

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