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The Atomic Iodine Laser

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Sandia Laboratories

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THE ATOMIC LINE LASER

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Introduction

The atomic iodine photodissociation laser has been under intensive study for a number of years. The physics associated with this system is now well understood and it is possible to produce a 0.1 nsec (or longer) near-diffraction-limited laser pulse which can be amplified with negligible temporal distortion and little spatial deformation. The output of either a saturated or unsaturated amplifier consists of a high-fidelity near-diffraction-limited, energetic laser pulse.

The applicability of the iodine photodissociation laser to the inertial confinement fusion program is not limited by any laser physics problems. We now know that an iodine laser system can be built that will satisfy the requirements of a laser-fusion power plant, but its efficiency needs to be improved. For a 1% laser efficiency, a source with at least 10% electrical efficiency is required. Because of limitations imposed by the requirement of chemical regeneration, the pump pulse width must be about 1 μ sec or shorter. For EPR-1 the source must have a lifetime of at least 10^5 shots and for an operating power plant, the pump must have a 10^9 shot capability. Coupled with these pump requirements is the necessity that the working gas be regenerated from the chemical products produced (during laser operation) with an efficiency of at least several percent. The overall efficiency of a gas-laser system must also take account of the energy to circulate the gas as well as for the energy required to cool the laser and pump sources. The above issues are addressed in this document, which is intended to serve as an aid to the office of Laser Fusion in deciding the level of effort that it will assign to research and development of the iodine laser.

This report is divided into three chapters. Chapter 1 is a survey of the important areas affecting efficient laser operation and summarizes the findings of Chapter 2. Chapter 2 presents detailed discussions and evaluations pertinent to pumps, chemical regeneration, and other elements in the overall laser system. Chapter 3 briefly discusses those areas that require further work and the nature of the work required to complete the full-scale evaluation of the applicability of the iodine photodissociation laser to the inertial confinement program.

CHAPTER 1: Overview and Conclusions

The atomic iodine photodissociation laser is well-characterized as a high power laser system. The excited iodine atom can be produced with unity quantum efficiency by photolysis of various alkyl iodides. The radiative lifetime is so long, 0.13 sec, that spontaneous emission contributes very little to the prepulse energy. The total lifetime is controlled by collisional deactivation, but is sufficiently long so that energy storage is not a problem. The stimulated emission cross section can be varied by adjusting the buffer gas pressure. This implies that the gain can be kept low to prevent self-oscillation of the amplifiers, but will not be so low that saturation fluxes are above the window damage limit. Although the atomic iodine laser transition is split by the hyperfine interaction, energy extraction at reasonable buffer gas pressures is efficient. The wavelength of the laser, 1.315 μm , is such that conventional high quality optics can be used, and the option of using a metal last mirror is available. High energy and short pulse production have been demonstrated at several laboratories. In addition the beam quality of a highly saturated output pulse has been shown to be excellent.

In considering the beam quality of a system for possible use as an ICF driver, one has to be concerned with saturation and non-linear effects, and with turbulence due to flow and mixing of the gases. In experiments conducted at Sandia and in theoretical modeling using a two-dimensional pulse propagation code, it has been shown that anomalous dispersion effects associated with saturation in an amplifier do not degrade the focusability of the atomic iodine laser. Calculations also indicate that spatial variations in the index of refraction caused by imperfect gas mixing and velocity fluctuations in the turbulent gas will result in less than 10% loss to scattered light even for large amplifiers.

Present atomic iodine laser systems are most commonly pumped by linear xenon flashlamps. Such pump sources have yielded iodine laser systems with at best 0.5% electrical efficiency. Many potential pump sources have been considered as alternates to xenon flashlamps but few can be projected to yield efficiencies greater than 1%. One attractive candidate is the metal-doped xenon flashlamp which has been shown to improve the efficiency by a factor of 2 - 3. However, technological problems exist with lamp lifetime and enhancement for short pulsewidths, which are required to minimize chemical

regeneration problems. Calculations indicate that pumping the iodine laser with the 254 nm band system of HgXe could result in a laser electrical efficiency of $\approx 3\%$; however, the appropriate experiments have as yet not been performed. A promising pumping scheme for the iodine laser involves chemical generation of $\text{O}_2(^1\Delta)$ which then dissociates I_2 and excites iodine atoms via energy transfer. This technique has been used to generate a cw iodine laser but must be investigated under pulsed extraction conditions.

In an atomic iodine laser based on photolysis of an alkyl iodide, RI , a portion of the starting material is converted to I_2 and R_2 . For an ICF driver, these materials must be used to regenerate RI . Two schemes appear to have sufficiently high projected efficiencies to warrant experimental investigation. Both schemes involve selective dissociation of R_2 followed by reaction of R with I_2 or I to produce RI . The two processes involve dissociation of R_2 via absorption of multiple CO_2 laser photons and dissociation of R_2 in a RF discharge. Current experimental results indicate both processes are highly selective in dissociating R_2 . The multiple photon technique has a measured efficiency of 2.5% for C_2F_6 which is comparable to the theoretical estimate of 3.5% for C_2F_6 . Larger R_2 have higher theoretical efficiencies.

The energy cost for flowing the laser gas, cooling the laser gas and cooling the laser head have been estimated for a conceptual design of a flow loop which has several parameters. It was found that an overall figure of merit* for this phase of the laser system was ≈ 1 for a moderate size amplifier using He as the buffer gas. The figure of merit decreases if Ar is used as the buffer gas and decreases with increasing laser output energy from a given module. However, it does not appear that the flow-cycle efficiency will limit the overall system efficiency.

As discussed above, an iodine laser module consists of several inter-related components which together determine the laser system's size and efficiency. A schematic representation of these components and their relationships is presented in Fig. 1, along with a table containing estimates of potentially realizable efficiencies for each item. The quantitative bases for these estimates are thoroughly documented in Chapter 2 of this report.

* The figure of merit for a particular step in the overall laser cycle is simply the ratio of the laser output energy to the energy required to carry out that step.

COMPONENTS OF AN IODINE LASER MODULE

Module Component	Percentage Efficiency or Figure of Merit
Power conditioning for pump	80 %
Fluorescence (intrinsic)	5 - 40 %
Fluorescence coupling	80 %
Extraction quantum	13 %
Laser electrical efficiency	0.42 - 3.3 %
Pump flow f.o.m.*	>1
Laser flow f.o.m.*	>1
Chemical regeneration f.o.m.**	0.03 - 0.10
Total module efficiency	0.36 - 2.4 %

* figure of merit, see text

Schematic diagram of the components which affect the efficiency of an iodine laser module.

Figure 1

It is evident from this table that the two subsystem components which impact the total system efficiency most heavily are the system electrical efficiency and the chemical regeneration efficiency. Significant improvements in electrical efficiency can be expected only through the discovery and development of pumps with high intrinsic efficiencies. The limiting chemical regeneration efficiency depends in a complex way on several variables, including the photodissociation kinetics of large perfluoroalkylhydrides and the temporal and wavelength characteristics of the pump source.

The tradeoff between the system electrical efficiency and the chemical regeneration efficiency in their influence on the total system efficiency is shown in Fig. 2. As a specific example, consider a system with 1.5% electrical efficiency. Then, a chemical regeneration efficiency γ of 1.2% is sufficient to obtain an overall system efficiency of 1.0%. Effort expended in doubling the chemical regeneration efficiency to 2.4% would raise the total system efficiency to 1.2%, whereas doubling the electrical efficiency to 3% increases the total system efficiency to 1.5%. Our present experimental data on element recovery indicates that 5% efficiency may be currently obtainable. If so, the pump source becomes the critical efficiency-limiting component.

Figure 3 shows the variation in total system efficiency with size of the laser module. As the module size exceeds 10^5 Joules of extractable energy, the cost of flowing the laser medium through the module begins to affect the efficiency of the system. The use of helium rather than argon as a carrier gas reduces the flow cost for a given module size, although, the total system pressure must be increased 30%.

The relationships among the aperture H , length L , and extractable energy density δ of a laser module of extractable energy E are given in Fig. 4. In each case, the parameter scaling is given for two values of the flux loading on the laser-module output window, 5 J/cm² and 20 J/cm². Note that for a module of fixed extractable energy, the length increases with increased flux loading of the output window, whereas, the height decreases. This particular dependence of L on flux loading is a direct consequence of the requirement of uniform photodissociation across the aperture of the module. The maximum module length is probably limited to < 50 meters because of distortion and scattering of the laser beam by medium inhomogeneities. Consequently, a laser system with extractable energy density of ≈ 1 J/kJ requires modules of ≤ 50 kJ extractable energy. Although rather long (50 meters), these modules have a modest 1 m² aperture.

TOTAL MODULE EFFICIENCY (INCLUDING FLOW)
-TRADEOFF BETWEEN ELECTRICAL AND
CHEMICAL REGENERATION EFFICIENCY

Contours of constant total efficiency for a defined laminar flow function of the electrical efficiency and the chemical regeneration efficiency. The energy cost (kWh/m³) for the base cell plus pump flow was assumed to be 0.01.

Figure 11

EFFICIENCY SCALING FOR AN IODINE LASER MODULE

Total efficiency for an iodine laser module as a function of extractable energy. The decrease in efficiency for large modules is a consequence of the increased energy cost of flowing the laser medium.

Figure 3

SCALING OF AN IODINE LASER MODULE

It is not possible to determine the efficiency of the system as a whole until the first two efficiency problems are solved. Pumping should be designed to be efficient and the system should be processing and flowing. If these two problems are solved, the two which limit overall efficiency are optical pumping and chemical conversion efficiency. It is noted that in the values of the efficiency given in Table I, there is a large overall system efficiency of 10.7% which is a very low value and that the value of the optical efficiency is even lower at 0.7%. The availability of any iodine is purely a chemical matter. Iodine laser is also discussed in this report, and the projected overall efficiency is approximately 6%.

The atomic iodine laser system is an attractive candidate for the future in the high efficiency fusion program based on its physical performance characteristics. The limiting factor in the overall system efficiency will be a combination of low efficiency in the pumping and chemical conversion steps. Current experiments in the area of optical pumping and laser processing and more work is needed in this area. Laser optical pumping cannot be ruled out and several schemes should be more fully evaluated. In addition, the complete evaluation of a pulsed iodine laser as a candidate for the fusion of iodine system must be performed.

2.1.2. Efficiency

Of course, the most important factor in the evaluation of the atomic iodine laser is its efficiency. The efficiency of the laser is defined as the ratio of the laser output power to the input power. The input power is the power of the pump source, and the output power is the power of the laser beam. The efficiency of the laser is a function of the pump power, the laser transition, and the laser cavity. The efficiency of the laser is typically in the range of 10% to 20%.

In this work, the efficiency of the atomic iodine laser is evaluated by using the following procedure. First, the laser transition is described with reference to the literature. The laser transition is described by the laser transition energy, the laser transition dipole moment, and the laser transition frequency. The laser transition energy is the energy difference between the upper and lower laser states. The laser transition dipole moment is the dipole moment of the laser transition. The laser transition frequency is the frequency of the laser transition. The laser transition energy, the laser transition dipole moment, and the laser transition frequency are used to calculate the laser output power. The laser output power is the power of the laser beam. The laser output power is a function of the pump power, the laser transition, and the laser cavity. The laser output power is typically in the range of 10% to 20%.

2.2. Individual Characterization of the Atomic Iodine Laser

In this section several physical characteristics of the atomic iodine laser are described. First, the production mechanism to populate the upper laser state is described. This is followed by a discussion of the kinetics of the atomic iodine states. Finally, some details of the laser transition itself are presented.

2.2.1. Production of the Upper Laser State

The upper laser state of the atomic iodine laser is the $5^2P_{1/2}$ manifold. This state is typically produced by the ultraviolet photolysis of iodine-bearing compounds. The carbon-iodine bond is excited by the absorption of

excited state is repulsive, the iodine atom dissociates from the molecule with conversion of it to the $5^2P_{1/2}$ ground state and probability 100% to the $5^2P_{1/2}$ ground state. The first comparison to produce iodine atoms was by H. I. and H. I.¹ while the mechanism of each different iodine atom production was not to be neglected. The iodine atom production was not to be neglected. The reason for this is that the rate of the reaction is very small. The other reason is that the reaction of iodine results in a lower dissociation rate of the $5^2P_{1/2}$ iodine state of the parent molecule.²

The width of the absorption band changes from 20 nm up to 10 nm in $^{127}\text{I}_2$ for various parent molecules. The central wavelength is 590 nm in $^{127}\text{I}_2$ and results in the parent molecule. Both $^{127}\text{I}_2$ and $^{129}\text{I}_2$ seem to be the most efficient source of excited iodine. Not only in the central wavelength of the absorption band for these compounds longer, but the width of the absorption band is broader as well. The same-broadening parent molecular may result in the more efficient production of excited iodine,³ but they suffer from quite large dissociation rates.⁴

2.2. Kinetics of the atomic iodine factor

A simplified picture of the energy levels in the atomic iodine system is shown in Fig. 5. A more detailed description can be found in Ref. 7. The upper $5^2P_{1/2}$ state is split by the hyperfine interaction into two levels and the lower $5^2P_{3/2}$ state is split into four levels. Since the chemical kinetics of the atomic iodine laser have been analyzed in detail elsewhere,⁸ only the kinetics involving transitions between the atomic levels will be discussed here.

Transitions between the hyperfine levels in the $5^2P_{1/2}$ state are forbidden for a van der Waals collision.^{9,10} The dominant relaxation mechanism is resonant exchange in a collision between a $5^2P_{1/2}$ iodine atom and a $5^2P_{3/2}$ iodine atom. Since the atomic iodine concentration is usually not very high under most conditions, the hyperfine levels in the $5^2P_{1/2}$ manifold can be considered not to relax between each other on any relevant time scale.

A typical atomic iodine laser oscillator lases only on the strongest transition, i.e., the $F = 3 \rightarrow F = 4$ transition. Since the $5^2P_{1/2}$ levels do

1.315μ

Hyperfine structure of the atomic iodine

$5^2P_{1/2} \rightarrow 5^2P_{3/2}$ laser transition.

Figure 5

the laser, to the extent that how to extract energy from the $5^2P_{1/2}$ level. The possible solution is to use an external magnetic field to split the $F=1$ and $F=2$ levels of the $F=2 \rightarrow 1$ transition. The use of single-line oscillators whose outputs are sized out also is open. The technique will well be allowed to tune the oscillator output across the hyperfine transitions.¹² Another possibility is to pump ions across the linewidth sufficiently so that the hyperfine transitions overlap. If a single oscillator frequency could efficiently extract energy from both energy storage levels, the hyperfine splitting is a fraction of a wave number, and several thousand Torr of buffer gas is sufficient to broaden the transitions into a single transition.^{7,12-14} The saturation flux under these conditions is still only a few J/cm^2 . Even in the case for which a two-line oscillator would be used (i.e., low buffer gas pressure), pressure broadening is the dominant broadening mechanism. The line is thus pressure-broadened, and the entire line profile can be extracted without spectral hole-burning.

Non-radiative transitions between the $5^2P_{1/2}$ state and $5^1P_{1/2}$ state can occur in a collision with an excited iodine atom and a parent molecule.² Products of the photolysis, especially radicals, can also cause collisional deactivation.¹⁵ However, in the case where the parent molecule is a perfluorinated alkyl iodide such as $\text{C}_6\text{F}_5\text{I}$, collisional deactivation does not noticeably decrease the excited state population density during pumping. Deactivation by a collision with a buffer gas atom has an impact vanishingly small rate.²

The hyperfine levels of the $5^2P_{3/2}$ ground state can relax among themselves by a van der Waals collision with a buffer gas atom.¹⁶ Hence, relaxation is quite rapid and in fact, it is generally accepted that this process accounts for most of the homogeneous broadening of the transition.¹⁰ The relaxation rate ν is related to the linewidth $\Delta\nu$ of the transition by the formula $\nu = 2\pi \Delta\nu$. Thus, measurements of the pressure broadening can determine the lower-state relaxation rate. For 1 Torr argon $\nu \approx 35 \times 10^6 \text{ sec}^{-1}$. The value for helium is approximately $2/3$ as large. Under most conditions in an atomic iodine laser system, the hyperfine levels in the $5^2P_{3/2}$ state relax rapidly during the laser pulse.

2.2.3. Iodine Laser

Wavelength - The wavelength of the $\text{I}^{\text{I}} + \text{I}^{\text{II}}$ transition is 1.315 μm . Another advantage that interaction of the laser with the target will be better than for longer wavelength lasers is penetration of seawater (optical). There are studies that show that the laser beam can penetrate directly up to 100 m in the sea. If the laser wavelength is short enough so that conventional near IR-visible filters can be used. These materials are in general at very narrow angles of transmission and development usually exhibit excellent optical quality and resistance to damage. Finally, since the wavelength is greater than 700 nm, the eye is safe from retinal damage is avoidable. This may prove critical in the design of the last optical surface in an ICF power section, where neutron fluxes might damage dielectric surfaces.

Excited State Total Lifetime - The radiative lifetime of the atomic iodine laser transition is ~ 0.1 sec.¹⁶⁻¹⁸ The total lifetime is controlled by collisional and diffusion to the walls. Under most circumstances the total lifetime is > 10 msec, and since pump pulse durations are only a few msec, energy storage is no problem. Also, since the radiative lifetime is so long, spontaneous emission contributes very little to the prepulse energy.

Stimulated Emission Cross-Section - By adjusting the buffer gas pressure, the stimulated emission cross-section σ can be adjusted from $< 10^{-14} \text{ cm}^2$ to $> 10^{-12} \text{ cm}^2$. This is exactly the range of interest for an ICF laser driver, since the following three criteria can be simultaneously satisfied. The small signal gain can be kept small enough to prevent self-oscillation of the amplifiers ($n\ell < 1$), and there can be sufficient energy storage ($n\ell > 10 \text{ J/cm}^2$) to reach saturation fluxes which are below the damage flux of optical components ($h\nu/b\tau < \text{several J/cm}^2$). Here n is the excited state density, ℓ the amplifier length, and b the degeneracy factor of the transition. The adjustability of the stimulated emission cross section is one of the strongest advantages of the atomic iodine laser as an ICF laser driver.

Pulsewidth - The pulsedwidth can be continuously tuned from ~ 100 psec to several tens of msec. Free inductive decay,¹⁹ mode-locking,²⁰⁻²² and gain-switching (or Q-switching)²³ can all be used to produce a wide range of pulsedwidths. The transition linewidth can be adjusted from 250 MHz to several tens of GHz by changing the pressure of the buffer gas. This allows bandwidth-limited pulsedwidths for a wide range of pulsedwidths, thereby improving

and the pump intensity of the tapered amplifier. Amplification will be achieved by allowing to tailor the pulse envelope for optimal transfer of the gain.

Heavy Extraction - Heavily saturated operation of the laser is possible at the expense of the non-linear index of saturation of the gain medium. In a heavily saturated medium, the small-signal gain is reduced to a point at which the population without pumping can provide a net gain. Recent measurements² of the beam cross section of the present device indicate that stimulated beam conversion likewise will be unimportant. Also, the saturation flux is typically a few J/cm^2 or less, so that saturation fluxes can be obtained before optical component damage levels are reached. Even so, the laser can be highly saturated, the stored energy can be extracted efficiently. The relative beam cross of the input and output beams can increase up to 10^4 of the stored energy to be extracted, provided that efficient heavy extraction is maintained.

All of the characteristics of the laser transition by itself are considered viable candidates appear to be favorable for application to a laser driver for an ICF reactor. The slight amplification of the small-signal gain of the $^{127}\text{I}_{1/2}$ upper state should pose no technical problem for efficient extraction of the excited state energy, and the production of pulses with excellent temporal and spectral properties is a foregone conclusion.

2.3. Beam Quality of Iodine Laser

The beam quality of the iodine laser system can be predicted with considerable confidence since the work on Gail-1 has confirmed the correctness of the theory used for the description of amplified pulses. These predictions relate to the pulse distortions produced in the amplification process itself. A further source of distortion in a repetitively pulsed system is the nonuniform character of the medium due to turbulence and to mixing of gases. These can be designed out of the system with the proper engineering, once the cause of the effects are identified. The next sections identify and analyze these problems and show that they are not severe obstacles to the development of a large repetitively pulsed iodine laser system.

2.3.1. Anomalous Dispersion

One of the primary objectives of the Sandia atomic iodine laser program was to compare the experimentally-determined beam quality of an amplified

Based on the above information, and of course after several discussions, it was decided that the first major design effort would be to design a small signal amplifier that would be "pre-amplified" and it was found that the design of a small signal amplifier produced in the current MAIL-1 system was not the best design and would make under the beam profile. This small signal amplifier would be an interstage try and had a nominal effect on the beam profile. The design of a small signal amplifier system we can summarize the first design of a small signal amplifier. First of all, pre-amplification of the small signal amplifier under small signal conditions (no saturation) will be performed at a bandwidth will not distort or defocus the pulse. Second, the amplifier saturation required for efficiency in the final stages of amplification will reduce noticeable phase shifts across most of the pulse duration. Third, we will use a small focusing region near the amplifier wall, but the energy content is low. Third, if intermediate amplifier stages are a consideration, spatial filters would be desirable to restore the pulse to near-diffraction-limited quality. The spatial filters should be used to beam since they would only be transmitting a few tens of Joules of energy.

2.3.2 Turbulent Gas Flow

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It is appropriate to determine the effect of both these random factors on the propagation of a pulse through a gain medium. If the correlation length l_c is less than transverse dimension of the resonator, and is much smaller than the local waist w , the coherent beam acts as though it is scattering through a medium with an extinction coefficient,

$$2\alpha = 2k^2 \langle \Delta n^2 \rangle. \quad (1)$$

In this equation, $k = 2\pi/\lambda$, $\int_0^L \alpha(r) dr$, $\alpha(r)$ is the correlation function of the index variation defined as $\langle \Delta n(r) \Delta n(0) \rangle / \langle \Delta n^2 \rangle$, and $\langle \Delta n^2 \rangle$ is the mean square fluctuation of the refractive index. The scattered part of the pulse emerges as an incoherent beam of increased divergence, typically of angular spread $2''(2l)$ as opposed to $2''(kw)$ for the unscattered part of the beam. Since l will be small compared to w , we must assume that the scattered radiation is lost from the system. We assume that a 10% loss to scattered light is tolerable inside each amplifier stage; this is used to set a bound on $\overline{\Delta n}$:

$$\overline{\Delta n} < (10 \cdot 2k^2 l L)^{-1/2}. \quad (2)$$

L is the length of the amplifier stage. We now show that this bound is easily satisfied by our projected iodine laser design.

The turbulence scale size l is estimated to be of whatever dimension characterizes the separation of the elements of the flow straightener system at the entrance to the laser cavity. Typically, we take this to be 1 cm and have the bound:

$$\overline{\Delta n} < 1.48 \times 10^{-7} \left(\frac{1 \text{ cm}}{L} \right)^{1/2} \left(\frac{10 \text{ Torr}}{L} \right)^{1/2}. \quad (3)$$

We have chosen a typical amplifier length of 10 m, but retained the scaling with L and L . The relation of $\overline{\Delta n}$ to density variation can be evaluated from the molar refractivity at standard temperature and pressure (STP):²⁸

$$n = 1 + \frac{\rho}{\rho_{\text{STP}}} \beta_{\text{STP}} \quad (4a)$$

$$\overline{\Delta n} = \overline{\Delta \rho / \rho} \frac{\rho}{\rho_{\text{STP}}} \beta_{\text{STP}}. \quad (4b)$$

ρ is the mean density, ρ_{STP} is the density at STP, and β_{STP} is a constant. The refractive index of our laser system will be dominated by about 5000 Torr

the mean velocity $\bar{u} = \frac{1}{L} \int_0^L u \, dx$ and the mean square velocity $\overline{u^2} = \frac{1}{L} \int_0^L u^2 \, dx$ and in terms of the turbulence intensity fluctuation:

$$\overline{u^2} = \bar{u}^2 + \overline{u'^2} \quad (66)$$

$$\Delta \rho / \rho = \left(\frac{\bar{u}'}{\bar{u}} \right)^2 \left(\frac{10m}{L} \right)^2 \quad (67)$$

The fluctuation in density $\Delta \rho$ due to turbulent flow and density variations in the gas is characterized by $\Delta \rho / \rho$ and is the same as $\Delta \rho / \rho$ in the case of a laminar flow.

$$\Delta \rho / \rho = \overline{u'^2} / \bar{u}^2 \left(\frac{10m}{L} \right)^2 \quad (68)$$

where $\overline{u'^2}$ is the root mean square of the fluctuation velocity u' and is 22.4 m/s for $Re = 10^5$. The bounds on density fluctuations are converted to velocity relations,

$$u' / \bar{u} = \overline{u'^2} / \bar{u}^2 = 25 \left(\frac{L}{5} \right)^{\frac{1}{2}} \left(\frac{10m}{L} \right)^{\frac{1}{2}} \quad (69)$$

$$u' / \bar{u} < 0.22 \left(\frac{10m}{L} \right)^{\frac{1}{2}} \left(\frac{1cm}{L} \right)^{\frac{1}{2}} \left(\frac{10m}{L} \right)^{\frac{1}{2}} \quad (70)$$

In light of the expected lack of growth of turbulence scale size across the transverse dimension of the laser cavity²⁹ and also in light of the relative ease of maintaining a flow with $u' / \bar{u}$ as low as 0.02,³⁰ we feel that gas turbulence presents no problem for the large scale iodine laser design. The only required engineering feature is the flow straightener which adds a small amount to the circulation power requirement.

2.1.1. Temperature and Gas Mixing

Temperature fluctuations in the gas laser medium cause density variations which scatter the beam. If these temperature fluctuations are distributed randomly and on a scale that is small compared to the beam diameter, the random scattering theory of Sutton²⁷ may be used to set bounds on the allowable temperature nonuniformity. In the previous section we derived

$$\Delta \rho / \rho < 2.8 \times 10^{-5} \left(\frac{1cm}{L} \right)^{\frac{1}{2}} \left(\frac{10m}{L} \right)^{\frac{1}{2}} \quad (8)$$

which may be expected to be just above room temperature (300 K).

$$\Delta T \approx .004 \times \left(\frac{1 \text{ atm}}{\text{cm}} \right)^2 \left(\frac{1 \text{ cm}}{\text{cm}} \right) \quad (9)$$

This result is not as strict as it appears since all temperature fluctuations are expected to relax away before entering the laser channel except those generated in the flow straightener upon entering. Since the passage of the gas through the flow-straightener honeycomb is analogous to a Joule-Thomson expansion with a pressure drop on the order of 10 Torr, we estimate that the temperature profiles introduced into the cavity have a scale size on the order of the straightener separation and a variation on the order of $(10 \text{ Torr} \times 11.47 \text{ K/atm}) / (7.6 \text{ Torr/atm}) = .005 \text{ K}^{11}$. These are the only temperature fluctuations that should be considered in the random scattering calculation.

Another kind of temperature fluctuation arises from the upstream damping of acoustic waves. The heat release of the weak acoustic waves present in the iodine laser is minimal. Previous studies¹² have concluded that proper acoustic engineering can avoid this problem.

The problem of mixing uniformity arises because the depleted iodine-bearing fluorocarbon must be replenished before recycling the laser gas medium. If the optical cavity contains randomly distributed unmixed components the refractive index is affected much the same way as by temperature fluctuations. The refractive index of a gas mixture is given by

$$n = 1 + \sum_i \frac{\rho_i}{\rho_{\text{STP}}} \beta_{\text{STP}}^i \quad (10)$$

where i denotes species and the relevant refractive index constants for our system are¹³

$$\beta_{\text{STP}}^{\text{Ar}} = .000281$$

$$\beta_{\text{STP}}^{\text{C}_3\text{F}_7\text{I}} = .001835$$

$$\beta_{\text{STP}}^{\text{I}_2} = .001869$$

$$\beta_{\text{STP}}^{\text{C}_6\text{F}_{14}} = .001800.$$

$$\frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \frac{e^{-x^2/2}}{x} dx = 0$$

$$\frac{1}{x} = \left(\frac{x-1}{x} \right) + \left(\frac{1}{x} \right)$$

[illegible]

U.S. DEPARTMENT OF THE ARMY, MEDICAL SERVICE

Many different techniques have been successful in the "hot cathode" excitation of excited iodine atoms for laser action. In the following sections, we will examine each of these methods in detail from the point of view of its applicability to the Laser Fusion Program. For this application, cathode and ion sources must have high electrical efficiency, high excitation rates of a few Hz, high reliability, and scalability to high power levels. Therefore, these attributes apply to all potential fusion laser drivers, not just to the one for the atomic iodine laser.

3.3.3. Hydrolytic Sources

The most common technique for producing excited atomic iodine is photo-dissociation of a suitable iodine-bearing compound, e.g., $\text{I}_2\text{PF}_6\text{I}$, at the proper wavelength, typically a 40 nm wide band centered near 280 nm. Such a process is known to have near unit quantum efficiency for producing excited atomic iodine, but the energy efficiency η_E is rather low, since a 280 nm photon is being converted to one at 1.3 μm . In addition, the photodissociation technique requires a light source that is separate from the active laser medium. This requirement imposes a photon coupling efficiency η_c less than 100%.

the maximum efficiency of the energy stored in the laser beam by the extraction of the useful laser energy is to have the η_{ex} . As a consequence of these efficiency considerations, the plasma gain efficiency, the minimum practical source η_{pl} is given by

$$\eta_{\text{pl}} = \eta_{\text{ex}} \eta_{\text{p}}$$

where η_{p} is the minimum possible electrical plasma gain efficiency. For the laser and η_{ex} was chosen to be 0.5. Thus, the minimum merit for an acceptable photolytic driver must be at least 0.1 for a 1% electrical figure of merit. This requirement on the photolytic driver of high efficiency is perhaps the most stringent.

Discharge-Driven Photolytic Sources

Discharge-driven photolytic sources are the most extensively used for the atomic iodine laser. They share highly developed, yet relatively simple technology, and all but one discussed in this section are capable of very intense pulsed brightnesses on the order of 10^{16} W/cm^2 . Typically the discharge circuit consists of the photolytic source, an energy storage capacitor, and a switch such as a spark gap or an igniter to apply the capacitor voltage to the source. The simplicity of these components allows for a relatively long lifetime (i.e. number of usable shots) compared to most other types of photolytic sources.

Seven different discharge driven photolytic sources will be discussed in this section. Table I summarizes the important characteristics of these sources and is useful in comparing their relative attributes. The fluorescence efficiency mentioned in Table I is defined as the UV radiant energy in the atomic iodine laser pumpband divided by the capacitor bank energy.

Linear Xe Flashlamps. Flashlamps are extremely reliable pump sources and have a very well established technological base. They are capable of producing intense pump pulses of duration as short as 1-sec.³³ The lamp lifetime is a function of how close to the explosion limit the lamps are driven,³⁴ but lifetimes in excess of 10^5 shots are reasonable. The material from which the wall is constructed can also influence the lamp lifetime, and linear xenon flashlamps with a lifetime of 10^7 shots have been built.³⁵ The one shortcoming of flashlamps is that they have a fluorescence efficiency

1. The proposed radiant energy/pump source of choice is 10^{-4} W/cm² which is too low for use as a pump for an atomic iodine laser in a 100 Hz cavity.

Metal-doped Xenon Flashlamps. The addition of a metal to a xenon lamp to improve the UV efficiency of linear Xe flashlamps is a factor of 10^{-2} .⁴⁵ However, there is some question as to whether the enhancement can be used to short pulsewidth flash lamps.⁴² If the current enhances the cathode by creating a long tail to the light pulse. However, preliminary experiments using 10 ns the repeat current that enhancement may be still well on a short time scale with little tail.⁴² If this is the case, metal-doped xenon flashlamps might be a suitable pump source for the atomic iodine laser and the problem of efficiency may be solved. The question is answered in the lifetime (number of shots) of metal-doped lamps. The repeat time is minute out of the plasma volume. Two possible solutions to this problem are flashlamps with molybdenum electrodes being developed by IBM, Burlington⁴⁶ and insertion of the Te-impregnated matrix down the middle of the lamp. The matrix could be high impedance to avoid shorting out the discharge and would replenish Te lost from the plasma volume. Metal-doped Xe flashlamps are very promising as an efficient pump source, but much developmental work remains to be done before their applicability can be completely established.

Ablative Flashlamps. Ablative flashlamps⁴⁴⁻⁴⁵ are an intense UV light source based on the ablative excitation (and subsequent radiation) of the wall constituents, especially silicon, which has several UV emission lines. These lamps will probably never have an acceptable efficiency, since much of the energy is consumed by the ablation process. Also, they have an inherently limited lifetime, since $\sim 10^{-5}$ cm of wall material is removed on each shot. There is a possibility that an exotic wall material can be developed which would have a low ablation energy and good material recovery, as well as suitable optical and mechanical properties. As of now, however, ablative flashlamps cannot be considered as a serious candidate for an acceptable pump.

Coaxial Xenon Flashlamps. These flashlamps are a geometric variation of linear xenon flashlamps, using either an inner or outer coaxial return path.⁴⁶ They are very intense and capable of submicrosecond pulsewidths due to their low inductance. However, their efficiency is similar to that of a

 

Exploding wire. Exploding wires⁵⁰ have the advantage of delivering large amounts of energy into relatively small volumes. Unfortunately, they are no more efficient than xenon flashlamps, since they are basically a gas discharge. Also, the discharge is in the gain medium itself; a transparent envelope to separate the discharge from the gain medium would undoubtedly be destroyed by the ensuing pressure shock. This puts strict limitations on the choice of buffer gas so that arcing may be avoided. Also, the gas recovery problem is very serious for two reasons. Not only is there no filter to prevent photolysis of the VUV absorption band,⁴⁹ but the discharge itself violently dissociates the parent molecule. The additional problem of repetitive operation may possibly be solved, if the initial plasma channel is produced by a pre-ionizing laser or by a wick that can be drawn continuously through the tube. Nevertheless, in light of the low efficiency and gas recovery problems, exploding wires will probably not be acceptable as a pump for an iodine laser in an ICF power reactor.

Hg lamps. Quasi-cw Hg lamps have been considered as a pump source for the atomic iodine laser.¹¹ For cw operation these lamps have the very attractive fluorescence efficiency of 42%, and it is assumed that this efficiency would be maintained if the current had a pulsed component in addition to the cw component. Unfortunately, these lamps suffer from very low intensity. This creates two problems. The first is that of positioning a sufficient number of lamps around the gain medium. The second is that long pumping times (4-10 sec) are required to deposit sufficient energy. This makes the chemical recovery problem quite severe and also adds to the gain medium turbulence problem, since the gas has enough time to set up density gradients caused by non-uniform deposition of the flashlamp light.^{12,13} While there may be no satisfactory technological solution to the problems associated with low intensity, Hg lamps should not be discounted altogether. In light of their potential for very high efficiency. Also, due to relatively gentle pumping, Hg lamps are very reliable and should have a lifetime > 10,000 hours.

Summary. Metal-doped Xe flashlamps and surface discharges both show promise for development into a reliable intense UV source with a fluorescence efficiency $\approx 15\%$ in the atomic iodine laser pumpband. This would correspond to an electrical figure of merit of 0.015. Hg lamps already possess very high efficiency, but the serious problems associated with low intensity remain to be solved. The other discharge- driven pump sources do not seem suitable for use as a pump for an atomic iodine laser in an ICF power reactor.

2.1.1.2. Pump Sources Driven by Electron-Beam Accelerators or Electron-Beam-Sustained Discharges

Before discussing individual pump candidates that are driven by electron-beam accelerators or electron-beam-sustained discharges a few words on the reliability and scalability of the drivers themselves are in order. Since the requirements for electron-beam accelerators used by themselves are more stringent than those for accelerators used in conjunction with discharges, comments will be directed toward systems using direct electron-beam pumping with the understanding that they apply equally to electron-beam sustained systems.

The problem of reliability was also considered in the tests. Some deliver into a diode each month, following on the order of 10⁶ shots. This is a much smaller number with present technology and will leave the possibility of obtaining the energy that would be obtained in a diode in a very short time.

The problem of reliability is one that has been addressed in our larger pulsed-power system. An electron-beam driver⁵⁷ has been successfully operated at 10⁶ shots. The driver delivers 100 kV into the foil and the time delay and the number of pulses delivered by the electron-beam device can be varied over a wide range. The driver source will operate at or below this energy flux. This is difficult because foil loading appears to be the factor limiting reliability in these electron-beam devices at this time.

An electron-beam pulsed⁵⁷ generator, pump, trigger circuit and switch has been operated successfully for 1×10^6 shots. The individual components have been successfully tested⁵⁶ for 10⁶ shots. The question of whether a pulser can be put together with a diode and foil and be operated at 5 pps for 10⁷ shots without foil failure remains unanswered.

Xe-Br-Br₂. Using transverse electron-beam excitation Swingle⁵⁷ and co-workers pumped a mixture of 2 atmospheres of Ar, 25 Torr of Xe and 1 Torr of Br₂. They saw emission from two band systems of XeBr and one band system of Br₂. They defined a fluorescence efficiency, η_f , as the energy emitted by these band systems divided by the energy deposited in the gas mixture. They measured an η_f of 11%. An estimate of 50% overlap of this fluorescence with the pumpband of a typical iodine-bearing compound yields a net electrical efficiency for the atomic iodine laser, η_L , of 0.44%. Modification of the pumpband to absorb at longer wavelengths could bring this efficiency up to a maximum of 0.89%.

KrF. The maximum η_f measured^{58,59} for KrF to date is 9-10%. This was achieved using both direct electron-beam excitation and electron-beam sustained discharges. The η_L of an iodine laser pumped by such a system would be approximately 0.76%. In discussions with Avco-Everett⁵⁸ we were told that an η_f for the KrF laser of 10-12% might be achieved in an electron beam sustained discharge. This would raise the η_L of the 1.315 μ laser up to 0.89%.

Raman shifted KrF. It has been suggested that the wavelength of KrF, 248 nm, might lead to undesirable fragmentation of the iodine-bearing compound. This wavelength could be Raman shifted to the first Stokes of N_2 at 277 nm. The conversion efficiency is approximately 50%, giving a maximum η_L for the atomic iodine laser of 0.45%.

Cl_2/Br_2 . The ${}^2\Pi_g \rightarrow {}^2\Pi_u$ transitions of I_2 ⁶¹ and F_2 ⁶² in the ultraviolet have both been measured to have fluorescence efficiencies on the order of 30%. The analogous emission bands in Cl_2 and Br_2 , at 258 nm and 292 nm respectively, might be expected to provide efficient pump sources for an atomic iodine laser.

To this end we have measured the fluorescence efficiency of Cl_2 in an Ar/Cl_2 gas mixture using longitudinal electron beam excitation and found it to be 7.2% yielding an η_L of approximately 0.58%. Previous work⁶³ on the I_2 442 nm band system suggests that it might be possible to increase the fluorescence efficiency of the Cl_2 258 nm band system by using a chlorine-bearing compound with a higher electron attachment rate than Cl_2 . Experiments to determine this are now in progress.

The work done by Swingle⁵⁷ and co-workers on $\text{XeBr}-\text{Br}_2$ fluorescence suggests that the η_F for Br_2 could be as high as 11% if all the energy emitted by the XeBr band systems in $\text{Ar}/\text{Xe}/\text{Br}_2$ mixtures is channeled into the Br_2 band system in Ar/Br_2 mixtures. In this case, η_L could be as high as 0.89%.

D_2 . Electron beam excitation of Ne/D_2 mixtures showed⁶⁴ continuum emission from 1600 to 3500 Å. This emission has been attributed to the $a{}^1\Sigma_g \rightarrow b{}^3\Sigma_u$ transition of D_2 . An η_F of ~ 6% was measured for this system. Approximately one-third of this emission overlaps the pumpband of typical iodine-bearing compounds yielding an η_L of ~ 0.16%. By modifying the absorption band of the iodine-bearing compound we might double this efficiency to 0.32%.

HgXe . Theoretical calculations indicate^{65,66} that η_F for the 254 nm band system of HgXe could be as high as 40% when pumped by electron beam sustained discharges assuming that the excited state concentration is kept low. This is necessary if excited state collisions with electrons are to be kept from competing effectively with the radiative channel for removal of HgXe^* . It should be possible to satisfy these requirements if E/N in the discharge is approximately 3×10^{-16} V-cm² and the fraction of HgXe^*

from the laser of CF_3I is the excitation of the CF_3I laser in this case also.

2.2.2. Broadening of the CF_3I Laser.

Consistent with their observation of the spectral hole in the fluorescence spectrum, the broadening of the laser spectrum in the presence of spectral overlap between the emission and the absorption profile of the parent compound and a mixture of excited iodine atoms. Earlier, possible pump sources with narrow emission spectrum were discussed and how the possibility of broadening the absorption profile of the parent molecule with an external source of increasing the spectral overlap. The possible solution to this problem is to broaden the absorption spectrum of the parent compound by adding vibrational energy to the molecule by thermal heating or absorption of IR photons in a vibrational mode of the molecule. In addition to increasing the chemical kinetic constant of the laser, broadening of the absorption spectrum of the parent molecule would result in a laser of smaller signal size.

2.2.2.1. CO_2 Laser Excitation of CF_3I Vibrations

Furmer *et al.*¹⁷ have investigated the broadening of the ultraviolet absorption band of CF_3I centered near 270 nm by absorption of CO_2 laser photons in the ν_1 vibrational mode of CF_3I at 9.6 μm . In their experiment, the absorption of 350 nm Kr^+ ion laser light by CF_3I in a 180 cm long cell was monitored for a pressure range of 0.03 to 3.0 Torr CF_3I . The absorption at 350 nm is zero for ground state CF_3I ; however, when the cell was irradiated with the R(14) line of the 9.6 μm CO_2 laser transition at a power level of $2.5 \times 10^7 \text{ W/cm}^2$, an induced absorption of $\sim 13\%$ was observed for 1 Torr CF_3I . This corresponded to an average absorption of 1-2 CO_2 laser photons per molecule in the cell in the ν_1 vibrational mode of CF_3I . The results of Furmer *et al.* suggest that broadening of the CF_3I absorption spectrum has occurred by vibrational excitation; however, no quantitative number is given to indicate the extent of the broadening. A knowledge of the modified absorption profile would be required to obtain a broadening factor from their results.

At Sandia, the investigation of the broadening of the CF_3I absorption spectrum by vibrational excitation has been extended to include measurements

SCHEMATIC DIAGRAM OF APPARATUS USED TO MONITOR BROADENING OF THE CF_3I ABSORPTION SPECTRUM

The experimental arrangement used for investigating the broadening of the CF_3I absorption spectrum.

Figure 1

of the sample and position of film of CF_3I , with and without CO_2 irradiation. The experimental arrangement is shown in Fig. 6. The experiment consisted of monitoring the transmission of xenon flashlamp light through a 5 cm cell with a spectrograph. A band of ~ 10 nm or 20 cm⁻¹ is observed in a single shot. For each a, central region and given experimental conditions, a spectrum was recorded for an empty cell, a given pressure of CF_3I without CO_2 irradiation, and the same pressure of CF_3I with CO_2 laser irradiation. Separate monitors determined the energy level of the flashlamp and CO_2 laser, a and b, c. The film spectra obtained were then scanned on a microdensitometer, yielding plots of the light transmitted through the cell for the various conditions. Figures 1 and 2 show typical microdensitometer traces for the flashlamp light transmitted through an empty cell and through 5 Torr CF_3I without and with CO_2 irradiation respectively. Using the film calibration curve, the absorption profile of CF_3I shown in the lower part of the figures can be constructed. The FWHM for the CF_3I without CO_2 irradiation is in good agreement with that determined more conventionally.⁶⁸ It should be noted that the broadened absorption profile with CO_2 irradiation is asymmetric with non-broadening to longer wavelength. This was observed in all broadened spectra.

The CF_3I pressure, the CO_2 laser line and the CO_2 laser power were varied to determine the optimum conditions for broadening the CF_3 absorption spectrum. The largest broadening observed was an increase in the FWHM by ~ 2.3 for 5 Torr CF_3I irradiated using the R(14) line of the 9.6 μm CO_2 laser transition at a power level of $\sim 1 \times 10^6$ W/cm². This broadening was accompanied by a reduction in the number density at line center of ~ 3 . It was anticipated that these two numbers should agree if those molecules shifted from line center moved to a useful absorption region. A check of the absorption of the CO_2 laser by the 5 Torr CF_3I before and after irradiation at 1×10^6 W/cm² indicated a 30% decomposition of CF_3I . This agreed with the missing molecules removed from line center but not showing up in the broadened profile. For the same conditions, a reduction in CO_2 laser power to 3×10^5 W/cm² decreased the decomposition to 11% but also decreased the broadening factor to ~ 1.4 . Continued reduction in CO_2 laser power resulted in further reduced broadening.

MICRODENSITOMETER RECORDS AND CALCULATED ABSORPTION
PROFILE FOR CF_3I WITHOUT CO_2 LASER IRRADIATION

Microdensitometer records of the flashlamp light transmitted through an empty cell, a 10 Torr CF_3I without CO_2 laser irradiation. Lower part of figure is absorption profile of CF_3I calculated from the two upper traces.

Figure 7

MICRODENSITOMETER RECORDS AND CALCULATED ABSORPTION
PROFILE FOR CF_3I WITH CO_2 LASER IRRADIATION

Microdensitometer records of the flashlamp light transmitted through a 10 mm cell of CF_3I with CO_2 laser irradiation. Lower curve is calculated absorption profile of CF_3I calculated from the two upper traces.

Figure 2

The above experiments involved absorption of the ν_1 vibrational mode of CF_3I . This band was selected by Barker et al. because of the high density of transitions, particularly in the region of the spectrum, which would facilitate multiphoton absorption processes leading to highly excited vibrational states. Because of the problems with decomposition arising when the ν_1 band of CF_3I is the $\nu_1 + \nu_3$ vibrational mode. Because the ν_1 band is so strongly saturated, a higher CF_3I pressure is required to deposit the same CO_2 laser energy thus decreasing the number of photons absorbed per molecule on the average. At 40 Torr CF_3I and a power level of $1 \times 10^6 \text{ W/cm}^2$ on the P(4) line of the 9.6 μm CO_2 laser transition, a broadening of 25% was observed. The broadening was found to increase as the ratio of absorbed photons/molecule increased. However, decomposition was also observed, although not quite as severe as with ν_1 .

Using ν_1 and $R(14)$, data were recorded at several delays of up to 100 μsec after CO_2 irradiation and found to be the same. Based on our estimate this should be sufficient time for vibrational equilibration of the CF_3I . This suggested that deposition of the same CO_2 laser energy over a longer time scale would result in the same broadening and might reduce the amount of decomposition because of the lower flux. By varying the flow rate of gases in the CO_2 laser, the pulse width of the laser was changed from ~ 100 nsec FWHM to ~ 2 μsec FWHM. With $R(14)$ and 5 Torr CF_3I , the same energy was deposited in the cell for the two pulse widths. In each case, the percentage of CF_3I decomposition was the same.

While broadening of the CF_3I absorption spectrum has been observed for many conditions, the problem of decomposition of CF_3I by the CO_2 laser was encountered. Unfortunately, it appears that the degree of broadening correlates directly with the degree of CF_3I decomposition. The decomposition of CF_3I by CO_2 laser photons limits the usefulness of this technique for two reasons. First, the decomposition products are CF_3 radicals and ground state iodine atoms. Thus, for 30% decomposition by the CO_2 laser and 25% photolysis by the flashlamp producing excited iodine atoms, the inversion is substantially reduced. Secondly, the decomposition of CF_3I increases the amount of material for chemical regeneration. If one could somehow avoid the decomposition problem, this technique would be an attractive way to improve the efficiency of the atomic iodine photodissociation laser.

... *Phragmites australis* ...

In an experiment to verify the above two points, FBI obtained copies of the entire correspondence between the defendant and the Bureau from the beginning to the end of the case, including letters, cables, and teletypes. FBI obtained a statement of all mail sent through the post office.

[illegible]

... the Sources for Producing Excited Iodine Atoms

There are several other possible mechanisms which can be used to produce excited iodine atoms. These possibilities include photodissociation of molecular iodine, electrical dissociation of alkyl iodides, and chemical excitation of excited iodine atoms. While extensive knowledge about these sources does not exist, the chemical pumping technique appears to offer the greatest potential for high efficiency. A brief discussion of each of these potential pumping schemes will follow.

2.4.2.1. Single Photon Dissociation of I_2

Photodissociation of molecular iodine by direct pumping to the B^3 state will result in the production of one excited iodine atom and one ground state atom. Because of the favorable degeneracy ratios in the atomic iodine system, this production ratio of I^*/I could result in stimulated emission. Recently, S. J. Davis⁶⁹ of the Air Force Weapons Laboratory produced an atomic iodine laser by selective pumping of I_2 with a tunable dye laser. He observed atomic iodine laser emission for dye laser pumping between ~490-525 nm. This technique suffers from the poor pump efficiency of dye

laser on the narrow excitation band if other pump sources are used. The energy storage capability of such a system is probably limited by the I_2 quantum rate of I^* unless a pump source capable of bleaching the I_2 could be used. For molecular iodine concentrations of about 1 Torr the decay time of excited iodine atoms is $\sim 1 \mu\text{sec}$.

2.4.3.2. Two Photon Dissociation of I_2

A method for pumping molecular iodine involving two photon dissociation has been proposed by C. K. Rhodes⁷⁰ of SRI International. This process has the potential advantage over the single photon dissociation that the dissociation occurs from a state which correlates at infinite separation of the atoms to two excited iodine atoms. However, the selective pumping region has not as yet been identified and the scheme would suffer from the same potential problems as the single photon method.

2.4.3.3. Electrical Dissociation of Alkyl Iodides

Transverse-discharge excitation of the atomic iodine laser has also been successfully employed as a pumping mechanism. Pleasance and Weaver⁷¹ observed atomic iodine laser emission when a mixture of 1% CF_3I in N_2 was excited in a UV-initiated pulse discharge between planar electrodes. However, because of certain experimental observations, some doubt arose that the laser may have been photolytically pumped by the UV-preionizer or the plasma light itself. Later, Wong and Beverly⁷² demonstrated that they had achieved atomic iodine laser emission by electron impact dissociation of $i\text{-C}_3\text{F}_7\text{I}$ in He. Although the efficiency in the above experiments was quite low, the main concern is the effect of the electric discharge on the chemical regeneration. Because of the large cross sections for ionization of the parent and dissociative electron attachment, the potential number of end products is much larger than by photodissociation in the proper pumpband. This would sufficiently hinder the chemical regeneration scheme that this process could not be considered even if the efficiency could be made as high as for photolytic pumping.

2.4.3.4. Free Electron Laser as a Pump for the Atomic Iodine Laser

The recent experimental demonstration of optical gain⁷³ and laser oscillation⁷⁴ in the free electron laser enhances the prospect that this device

may develop into a more efficient laser light source. There has been a substantial amount of theoretical effort on the free electron laser, and both quantum mechanical⁷⁵ and classical⁷⁶ descriptions give reasonable estimates of the small signal gain observed in Ref. 74.

There are two possible ways in which a free electron laser might be used to pump an iodine laser in an ICF reactor. The first is to operate the free electron laser at 253.7 nm and photo-dissociate perfluoroalkyl iodides such as PF_3I . Realistic estimates of the fluorescence efficiency of a free electron laser utilizing a storage ring to recirculate the electron beam pump current are $\sim 10^{-2}$ to 10^{-3} or lower.⁷⁷ This is equivalent to a figure of merit of $\sim 10^{-2}$ for the overall electrical efficiency.

The other possibility is to operate the free electron laser at 2500 nm and to photo-dissociate molecular I_2 . It has been shown that this wavelength can excite iodine into the $5^2\text{P}_{1/2}$ state and excite iodine atoms in the $5^2\text{P}_{3/2}$ state with a quantum efficiency of unity.⁷⁷ While the extractable stored energy is only $1/4$ as large as in the case where only excited states are produced, there are two offsetting advantages. First, the ratio of output photon energy to pump photon energy is almost twice as great as in the case of the photolysis of perfluoro-alkyl iodides. Second, the requirements for recycling the original parent are far simpler if the parent is I_2 . The overall figure of merit should be ~ 0.1 if the fluorescence efficiency of the free electron laser is 20% and if I_2 is used as the parent molecule. One complication is that the pulsewidth of the free electron laser must be ≤ 100 nsec, to avoid deactivation of the $5^2\text{P}_{1/2}$ iodine atoms by the I_2 . However, since electron accelerators and storage rings using klystrons as the RF power generation source are inherently pulsed devices,^{78,79} this constraint should not be severe. The added advantage of klystrons is that they are very efficient⁸⁰ and extremely reliable.

There is one potentially very serious problem that may limit the ultimate scalability of the free electron laser to very high power and simultaneously very high efficiency. Even at the saturation flux, the efficiency of the free electron laser is only a fraction of a percent for a single pass of the electron beam.⁸⁰ In order to obtain the 20% efficiency mentioned above, the beam must be reamplified and reinjected several times. As the electron beam pumps the optical radiation, not only does it lose energy, but the momentum distribution is broadened.^{78,80} It is essential that the initial narrow

momentum distribution be recovered before the electrons are reinjected. If the distribution cannot be recovered, the ultimate saturation flux of the free electron laser will undoubtedly be unacceptably low. The ability to recover the initial momentum distribution is currently under study.⁷ If the results prove favorable, then the free electron laser has a good chance of developing into an efficient and intense pump for the atomic iodine laser, which is a better laser medium for a good energy storage, nanosecond or picosecond pulsewidths, and large aperture.

2.4.3.5 Chemical Generation of Excited Iodine Atoms

Another interesting pumping mechanism for the atomic iodine laser is energy transfer from $O_2(^1L)$. This technique involves chemical generation of $O_2(^1L)$, the excitation energy of which is nearly resonant with the transition between excited iodine atoms and ground state iodine atoms. Thus, single energy transfer occurs from the $O_2(^1L)$ state to ground state atomic iodine producing the upper laser level.

Recently, a cw atomic iodine laser has been demonstrated at the Air Force Weapons Laboratory⁸¹ (AFWL) based on this technique. The $O_2(^1L)$ was generated by bubbling Cl_2 gas through basic hydrogen peroxide. Yields in excess of 40% $O_2(^1L)$ were achieved at 1 Torr delivery pressures. Yields as high as 80% are anticipated for a modified chemical generator design. The important chemistry involved in their system is shown in the following reactions:

The transfer of $O_2(^1L)$ to I is nearly gas kinetic with the rate-limiting process being the generation of $O_2(^1L)$ to be used in I_2 dissociation. The AFWL process is purely chemical in that ground state I is formed in reaction (II). However, it may be more practical for other situations to form I separately and inject it into the system.

To establish whether this type of device has any potential applications to an iodine laser system used for fusion applications. Preliminary calculations suggest that a number density of excited iodine atoms for useful energy output is achieved. The equilibrium between $I_2^{(1)}$ and $I_2^{(2)}$ is such that only about 1% of the total excitation energy is in the iodine at any given time. However, one can envision multiple extraction of the laser medium with re-establishment of the equilibrium between excitations so that the entire energy in $I_2^{(1)}$ could be recovered. A very attractive feature of such a laser system is that there is no pulsed power supply. Also, electrical energy will undoubtedly be required. It will be of the type involving low-capital expenditure and very high reliability.

The efficiency of such a device is determined on the energy required to regenerate the chemical reaction starting materials in the production of $O_2^{(1)}$. The balance equation for the chemical production of $O_2^{(1)}$ is given as:

Assuming an 80% production of $O_2^{(1)}$, and efficient extraction of energy, the thermodynamic upper limit on the efficiency would be about 13%. Regeneration of H_2O_2 can be accomplished from H_2 , O_2 and H_2O using an anthraquinone synthesis which consumes very little energy. Regeneration of the Cl_2 and H_2 required for the H_2O_2 synthesis involves electrolysis of HCl. Using the values for the commercial production of Cl_2 by electrolysis,⁸⁵ the overall laser efficiency is projected to be 6%. Since this efficiency should be compared to the combined electrical pumping efficiency and chemical regeneration of a photolytically pumped iodine laser, the technique looks interesting and deserves more attention.

2.5 Chemical Regeneration

2.5.1 Introduction

As is well known, the usual starting material employed in an iodine laser is a perfluoroalkyl iodide, RI. The operation of an iodine laser results in the conversion of some of the RI into R_2 and I_2 . Because RI is expensive, it is necessary to regenerate RI from R_2 and I_2 rather than to replace it by purchase of new material. Reference 8 and 26 present

calculations estimating the amount of H_2 formed as a function of relevant laser operating parameters and discuss the economic motivation for chemical regeneration. These reports describe in detail the theoretical basis for a regeneration scheme based on dissociation of H_2 via multiple photon absorption. In the literature and Reference 26 also suggests that chemical reactions in low frequency discharges might lead to efficient regeneration. The following material briefly summarizes the calculations showing the dependence of RI destruction on laser-operating parameters, discusses possible regeneration schemes, presents current results of ongoing regeneration experiments at Sandia and gives projected values of chemical regeneration figures of merit ("efficiencies") where appropriate.

When an amplifier using RI as a starting molecule is optically pumped, many excited iodine atoms I^* are produced. Extraction of optical energy converts most of the I^* population to ground state atoms, I . The rates of the various processes which take place in the mixture are discussed in References 6 and 26. The most important processes are

The competition and interplay between (VI), (VII), and the other processes of importance in an iodine amplifier have been treated numerically via computer solutions of coupled sets of differential chemical reaction rate equations.⁸ Figure 5 presents the dependence of the number of molecules of RI which are lost via (VII) per laser output photon on the rate constant for (VII) for several values of Δt , the optical pump pulse width.

Two features emerge clearly from Fig. 9. First, less RI is lost the shorter the pump pulse. This is because ground state atoms are present in appreciable concentration only after the completion of optical pumping and energy extraction. Before this time process (VII) can generate unwanted R_2 with little competition from (VI). The chemical regeneration problem is reduced by minimizing the time during which (VI) cannot compete with (VII), that is by minimizing pump pulse width. Second, the amount of destruction of RI is sensitive to k_{RR} , the rate constant for (VII). The chemical regeneration problem is reduced by minimizing k_{RR} . A well-designed iodine laser system will take into consideration these features but, as shown in

Effects of radical-radical association rate constant plotted only with k_{RR} a constant of starting material in a iodine laser cell. Laser pulse width = $10 \mu \text{ sec}$. See text for several comments of interest.

Figure 1

Reference 8, economic considerations dictate that chemical regeneration will still be required.

Finally, a simple, purely chemical procedure might be found to carry out the process

Unfortunately, the R_2 molecule, a perfluorocyclohexane, is very stable both thermodynamically and kinetically. The specific carbon-carbon bond holding the two R groups together must be attacked in order to make (VIII) proceed and there is no known chemical route for this attack. Before shelving the search for a purely chemical technique for regeneration, Sandia personnel discussed the problem with chemists of several disciplines in both industry and university. The unanimous response was that an efficient purely chemical scheme did not exist. It should be mentioned that destructive pyrolysis of R_2 has been discussed as the basis of a regeneration scheme^{9a} but that it does not lead to an efficient process because the pyrolysis step is not selective and not in the manner in which it attacks R_2 .

In the absence of any purely chemical procedure for carrying out (VIII), schemes involving physical steps have been pursued. The most promising physical step appears to be the dissociation of R_2

because the subsequent reaction of R with I_2 or I to form RI is spontaneous. Two physical schemes for carrying out (IX) are presently being investigated: dissociation of R_2 via multiple photon absorption in the CO_2 laser frequency region and dissociation of R_2 in continuous wave alternating current electrical discharges. Studies using each of these techniques are described below. A third approach which would also utilize a CO_2 laser was discussed briefly in Reference 8. This approach has not yet been investigated, but may prove interesting and useful in the future.

2.5.2 Dissociation via Multiple Photon Absorption

The absorption of a single infrared photon leaves a molecule vibrationally excited but stable. The absorption of a large number of such photons leaves a molecule sufficiently excited that it can dissociate if a large fraction of the energy finds its way into one bond. Reference 8 presents calculations of dissociation rates as a function of absorbed energy based

Simplified schematic of apparatus for spectroscopy and laser calibration system. The TEA CO₂ laser and the main attenuator, 75 cm path length cell, and the 5 cm path length cell are shown in the top right. The Mo mirror is shown in the bottom right. The diagram is a simplified schematic of the apparatus.

on a statistical theory for the rate at which energy migrates through the bands of a molecule and builds up in a particular one. The calculations in Reference 8 indicate that it should be possible to use a CO_2 laser to excite R_n in such a way that (IX) proceeds at an appreciable rate and that all other dissociation processes proceed at negligible rates. Based on the work in Reference 8, the experiments described below were initiated.

Effort has been concentrated on the simplest RI of interest, CF_3I , and the corresponding R_2 , C_2F_6 . The first step of the experimental work was determining the absorption strength of molecules of interest in the CO_2 laser frequency region. Figure 10 is a simplified schematic of the apparatus used for spectroscopic studies and for high intensity irradiations. The same laser is used in both kinds of studies so that there will be no change in the temporal behavior or frequency distribution from one experiment to the other.

It was determined that C_2F_6 absorbs in two different parts of the CO_2 μm ($00^\circ 1 - 02^\circ 0$) laser band. The ν_5 fundamental absorbs R(30) and lines of shorter wavelength. The $\nu_2 + \nu_{12}$ and $\nu_7 - \nu_{12}$ combinations absorb the lines P(30)-P(46). Detailed work focused on these two regions. Absorption strengths were determined for C_2F_6 , CF_3I , $\text{C}_2\text{F}_5\text{I}$, CF_3H , and $\text{C}_2\text{F}_5\text{H}$. Spectroscopic data were obtained at fluences of about 0.3 J/cm^2 (intensities of about $2 \times 10^5 \text{ W/cm}^2$). These values are low enough that no saturation effects should be important at the typical operating pressures of several Torr.

Table III reports absorption strengths in terms of the phenomenological, or effective, absorption cross section defined as

$$\sigma_{\text{eff}}(\text{cm}^2) = \left(\frac{1}{nL} \right) \ln \left(\frac{T_e}{T} \right) \quad (14)$$

where n is the total number density of the absorbing molecule, L is the absorption path length, and T_e and T are the fractional transmissions of the evacuated and filled cell, respectively. Data were taken at different pressures for different molecules in order to optimize signal/noise.

Even though the R branch lines are about 25 cm^{-1} to the red of the ν_5 band center of C_2F_6 , Table II indicates that the σ_{eff} values are larger in the R branch than in the P branch. For this reason, high intensity irradiations were undertaken in the R branch. The line chosen was R(36) because the laser fluence on lines with larger values of σ_{eff} was appreciably lower than

Table 11

Experimental results for $\sigma_{\text{eff}}^{(F+G)}$ in μbarn for $\sqrt{s} = 1.6$ GeV

Particle	$\sqrt{s}$ (GeV)	<u>Branch</u>			
		$\sigma_{\text{eff}}^{(F+G)}$ (μbarn)	$\sigma_{\text{eff}}^{(F+G)}$ (μbarn)	$\sigma_{\text{eff}}^{(F+G)}$ (μbarn)	$\sigma_{\text{eff}}^{(F+G)}$ (μbarn)
$\pi^+\pi^-$	1.6	6.1×10^{-11}	6.1×10^{-11}	1.1×10^{-10}	1.1×10^{-10}
$\pi^+\pi^0$	1.6	1.1×10^{-10}	1.0×10^{-10}	1.1×10^{-10}	1.1×10^{-10}
$\pi^+\pi^+\pi^-$	1.6	1.0×10^{-10}	1.1×10^{-10}	1.1×10^{-10}	1.0×10^{-10}
$\pi^+\pi^-\pi^0$	1.6	6.0×10^{-11}	1.1×10^{-10}	1.1×10^{-10}	1.1×10^{-10}
$\pi^+\pi^-\pi^+\pi^-$	1.6	1.1×10^{-10}	1.1×10^{-10}	1.1×10^{-10}	1.1×10^{-10}

Table 12

Experimental results for $\sigma_{\text{eff}}^{(F+G)}$ in μbarn for $\sqrt{s} = 1.6$ GeV

Particle	$\sqrt{s}$ (GeV)	<u>Branch</u>		
		$\sigma_{\text{eff}}^{(F+G)}$ (μbarn)	$\sigma_{\text{eff}}^{(F+G)}$ (μbarn)	$\sigma_{\text{eff}}^{(F+G)}$ (μbarn)
$\pi^+\pi^-$	1.6	6.1×10^{-11}	6.1×10^{-11}	6.6×10^{-11}
$\pi^+\pi^0$	1.6	7.0×10^{-11}	1.1×10^{-10}	1.1×10^{-10}
$\pi^+\pi^+\pi^-$	1.6	5.1×10^{-11}	1.1×10^{-10}	1.9×10^{-11}
$\pi^+\pi^-\pi^0$	1.6	6.1×10^{-11}	1.1×10^{-10}	1.2×10^{-10}
$\pi^+\pi^-\pi^+\pi^-$	1.6	7.0×10^{-11}	7.6×10^{-11}	6.1×10^{-11}

that for CF_3I . Table II shows that CF_3I absorbs much more strongly than CF_3H . Experiments in which CF_3I is regenerated from C_2F_6 and I_2 must therefore be carried out in such a way that the CF_3I product is never exposed to the laser radiation, lest it be decomposed. This will not be difficult in a well-designed commercial process, but it greatly complicates initial experiments. CF_3H absorbs appreciably less strongly than C_2F_6 and can be exposed to fairly high intensity laser radiation with little decomposition. This simplifies experimental design and is the reason that current work focuses on carrying out the process

via the intermediate step (IX) followed by reaction of CF_3 with H_2 or H . Most of the information obtained in this study will be directly transferable to the $\text{C}_2\text{F}_6/\text{I}_2$ system.

In order to determine the conditions appropriate for carrying out (X), various mixtures of C_2F_6 and H_2 have been subjected to pulsed CO_2 laser irradiation using the apparatus shown in Fig. 6. The R(36) line at $9.192 \mu\text{m}$ is used and the typical fluence is 6 J/cm^2 . The irradiation pulse width is approximately 10^{-7} sec. Infrared spectra of samples are obtained before and after irradiation using a commercial recording infrared spectrophotometer. In this way the nature and concentration of reaction products can be determined. It has been found that (X) proceeds, and that there is only one undesired by-product, C_2F_4 . It is not certain what the mechanism of C_2F_4 production is, but the most plausible one involves

followed by elimination of HF from $\text{C}_2\text{F}_5\text{H}^\dagger$. The symbol $\dagger$ denotes vibrational excitation. Unfortunately, there is no chemical kinetic information relevant to (XI) in the literature.

The overall reaction efficiency and the ratio of CF_3H production to C_2F_4 production have been determined for a number of $\text{C}_2\text{F}_6/\text{H}_2$ mixtures. The optimum pressure of C_2F_6 has been found to be 3 Torr. Figure 11 shows the dependence of the fractional consumption of C_2F_6 in the irradiated volume per laser pulse on hydrogen pressure for a C_2F_6 pressure of 3 Torr. In order to make inferences about possible mechanisms for multiple photon absorption, data were obtained with C_6H_{14} in place of H_2 and these are also shown in

Fractional consumption of C_2F_6 in irradiated volume per laser pulse as a function of p_x at fluence of 0.05 J/cm² per pulse. Line segments connecting data points have a slope of 1 at minimum $p_{C_2F_6}$ of 0 Torr.

Figure 11

$\text{CF}_3\text{H} / \text{C}_2\text{F}_4$ production ratio for 20 pulses of CO_2 R(36) radiation at fluence of $6\text{J}/\text{cm}^2$ per pulse. Line segments connecting data points have no theoretical significance $P_{\text{C}_2\text{F}_6} = 3$ Torr.

Figure 12

The present status of this work is that values of τ_h compatible with an overall system efficiency of 0.01 have been attained, assuming that the present work can be transferred directly to the C_2F_6/I_2 system. It is not known whether C_2F_4 will be a product in the C_2F_6/I_2 system. If it is, C_2F_4 can be fluorinated to C_2F_6 and recycled with some reduction in overall regeneration figure of merit. It must be emphasized that the present experimental conditions are probably not optimum and that appreciably larger values of τ_R may be attained in the near future.

2.5.3 Dissociation and Reaction in Electric Discharges

Another promising technique concerning the regeneration of RI compounds from R-R compounds involves the use of electric discharges. An example of fluorine compound synthesis using an electric discharge technique can be found in the work of R. J. Lagow and co-workers⁸⁵ who have studied the formation of a number of trifluoromethyl organometallic compounds. With certain variations, the technique has also been used for the synthesis of CF_3Cl from C_2F_6 and LiI_2 (ref. 86), and used in the synthesis of fluorinated polymeric compounds⁸⁷. While the technique appears promising for the regeneration of R-I compounds from R-R compounds, little work exists in the literature which would suggest the most efficient way to utilize this technique for regeneration of R-I compounds from R-R compounds nor is there enough input from the literature to estimate in a realistic way what the energy cost per R-I molecule would be for a large-scale regeneration facility.

The goals of this initial study are as follows: 1) to determine that it is possible to regenerate RI compounds of specific interest from R-R compounds using electrical discharge techniques; 2) to perform parametric studies concerning the optimization of formation efficiencies of the desired compounds; and 3) to utilize such information in the projection of a large facility for regeneration as required in a full-scale iodine laser system.

The electric discharge regeneration technique can be described as follows. An R-R compound such as C_2F_6 is passed through a chamber containing an electric discharge such that the C-C bond is broken to give CF_3 radicals. Because the C-F bond energy is considerably higher than the C-C bond energy⁸⁸ there can be many more C-C bonds broken in the discharge than C-F bonds. Thus depending upon the nature of the discharge, the gas pressure, and the flow rates involved, it is possible to produce CF_3 as the predominant radical

species formed in the discharge. Once CF_3 is formed, it can then be reacted with I_2 giving

and

where C_2F_6 and I_2 can act as the third body, M. The details of our studies concerning the utilization of electric discharge techniques are given below.

A block diagram of our initial setup to investigate the interaction of electric discharges with R-R compounds is shown in Fig. The system is designed so that gas can flow through the discharge tube at a given mass flow rate and at any independently controlled pressure by first setting the gas flow rate at the flow rate controller and then adjusting the pressure control valve at the exhaust end of the flow system. Two quadrupole mass spectrometers are utilized in this study. One spectrometer is located immediately downstream from the discharge so that species present in the discharge can be identified. The second spectrometer is located about 70 cm downstream from the discharge so that end-product analysis can be achieved. Although the primary gas flow is established at the flow rate controller, a second gas injection orifice has been incorporated just downstream from the discharge unit so that gas reactants can be injected into the flow just after radical formation occurs in the flow system. This secondary gas injection system is used in connection with gas titration experiments as an adjunct to the mass spectrometers in determining the kinds and amounts of radical species formed in the discharge. It is also utilized to allow radicals to react with a second gas downstream from the discharge rather than directly in the discharge and thus to prevent product breakup in the discharge.

Initial studies were carried out using a microwave discharge unit operating at 2.4 GHz with total power dissipation of up to 100 W. Results obtained have shown that when C_2F_6 is passed through such a discharge a permanent modification of this gas can take place. The mass spectrometer cracking pattern contains a peak at atomic mass position 119 and is identified as $C_2F_5^+$. The amplitude of this peak is proportional to the amount of C_2F_6 left in the flowing gas sample as it passes through the discharge. Studies of this mass peak as observed at the two mass spectrometer positions

Block diagram of a flow-on line only static ionization mass spectrometer (continued).

Figure 1

(Fig. 13) show that the parent of the C_2F_4 can be detected in the discharge (depending upon discharge power, gas flow rate and pressure) and the CF_3 radicals formed in the discharge can associate to re-form C_2F_4 downstream from the discharge. However, the compound C_2F_4 is also formed in the discharge and is readily identified by the parent peak at atomic mass position 100 as the $C_2F_4^+$ ion and atomic mass position 91 ($C_2F_3^+$ which are in + present as part of the C_2F_4 cracking pattern.

The relative yield of C_2F_4 as a function of discharge power is shown in Fig. 14 for a given C_2F_6 pressure and flow rate through the discharge. A comparison of the amount of C_2F_4 observed immediately downstream from the discharge and that observed at the mass spectrometer 70 cm. downstream from the discharge has shown that essentially all of this compound is formed in the discharge rather than forming as a secondary reaction product occurring downstream from the discharge. Since C_2F_4 can react in the discharge to form a long chain polymer, it is desirable to operate the electric discharge at a low power level where production of this species is minimized. Increasing the flow rate of C_2F_6 through the discharge for a given discharge power tends to reduce the C_2F_4 yield for flow rates in the range of 20 to 50 cm./sec. It appears likely that relatively high flow rates will be required in order to minimize production of C_2F_4 . The relative yield of C_2F_4 decreases with decreasing C_2F_6 pressure in the pressure range of 0.5 Torr to .002 Torr. As will be discussed below, the apparent yield of CF_3 radicals approaches a maximum at less than 0.1 Torr of C_2F_6 (for a relatively low power discharge and high flow rate) so that it is possible to obtain a low C_2F_4 production efficiency under conditions of maximum production efficiency for CF_3 radicals.

A summary of initial results regarding C_2F_4 production is as follows: the relative amount of C_2F_4 produced is minimized for low discharge power, high flow rate and low C_2F_6 pressure.

Some difficulty has been experienced in determining absolute yields of CF_3 radicals in C_2F_6 . Since the main cracking pattern peak observed for C_2F_6 occurs at atomic mass position 69, i.e., the CF_3^+ ion, detection of the parent CF_3 radical peak in the presence of C_2F_6 is difficult. In order to identify the CF_3 radical in the presence of C_2F_6 an attempt has been made to react CF_3 formed in the discharge with other gases to produce products which can be detected in the presence of C_2F_6 . Although I_2 might seem to be a logical

C_2F_4 YIELD WITH INCREASING
DISCHARGE POWER

Production of C_2F_4 as a function of microwave power.

Figure 14

choice as a reactant gas to produce CF_3I which can be detected in the presence of C_2F_6 , iodine has not been utilized in these initial studies because of mass spectrometer contamination problems involving this compound. However, a successful approach to identify CF_3 in the presence of C_2F_6 has involved the reaction of Cl_2 with the radicals produced in C_2F_6 . The CF_3 radical can react with Cl_2 as follows:

The cracking pattern of CF_3Cl contains peaks at atomic mass positions 85 and 87 corresponding to the CF_2Cl^+ ion as well as having a strong peak at atomic mass position 69 corresponding to the CF_3^+ ion. When Cl_2 is injected into the flow through the orifice immediately below the electric discharge (Fig. 13), peaks at atomic mass positions 85 and 87 do occur during the time C_2F_6 is passed through the electric discharge, but not when the discharge is turned off. The buildup of CF_3Cl as a function of C_2F_6 pressure (with excess Cl_2) is shown in Fig. 15 for the case where a 5W microwave discharge is used with a C_2F_6 flow rate of 100 cm/sec. It is seen from this figure that the absolute yield of CF_3Cl increases with C_2F_6 pressure until a C_2F_6 pressure of 0.04 Torr is reached. Higher C_2F_6 pressures do not result in higher absolute CF_3Cl yields. In Figure 16, the CF_3Cl yield as a function of Cl_2 injection pressure (where Cl_2 is injected just downstream from the discharge) is shown for a 5W discharge and 0.02 Torr of C_2F_6 flowing through the discharge at 100 cm/sec. It is seen from this figure that the CF_3Cl yield increases with increasing Cl_2 pressure until about 6 Torr of Cl_2 is being applied to the injection orifice. Adding more Cl_2 does not increase the CF_3Cl yield which implies that all of the available radicals produced in the discharges can react with Cl_2 if at least 6 Torr of Cl_2 is applied to the injection orifice. (This is equivalent to 0.02 Torr of Cl_2 within the flow system.)

Although absolute mass spectrometer calibrations concerning the amount of CF_3Cl actually produced in the system are not yet complete, an upper-limit estimation of the CF_3Cl yields can be obtained by observing the absolute amount of Cl_2 utilized in forming CF_3Cl . In Fig. 17 two curves are shown representing the mass 70 Cl_2^+ signal as a function of Cl_2 injection pressure in the flowing system for the case where C_2F_6 is flowing through the discharge at 100 cm/sec. The top curve was obtained for the case where the discharge is off. With no discharge, the Cl_2^+ mass peak increases linearly with

CF_3Cl YIELD WITH INCREASING C_2F_6
PRESSURE (EXCESS Cl_2 PRESENT)

The yield of CF_3Cl produced by injection of Cl_2 just downstream from the discharge in a flowing C_2F_6 gas stream at various pressures.

Figure 15

CF_3Cl YIELD WITH INCREASING
 Cl_2 PRESSURE

The yield of CF_3Cl produced in a stream of C_2F_6 passing through a 5-watt discharge as a function of Cl_2 pressure injected into the flow downstream from the discharge.

Figure 16

CL₂ SIGNAL AS A
FUNCTION OF CL₂
INJECTION PRESSURE

The effect of a 5-watt discharge on the downstream Cl₂ concentration due to interaction of Cl₂ with radicals produced in a C₂F₆ gas stream passing through the discharge.

Figure 17

increasing Cl_2 pressure in the flow. When the discharge is turned on (5W), the Cl_2^+ peak does not initially increase linearly with Cl_2 injection pressure because at the low Cl_2 pressures most of this gas is utilized in forming CF_3Cl . However, as shown from the lower curve, as the Cl_2 injection pressure is increased so that excess Cl_2 occurs in the flow system the Cl_2^+ signal becomes linear with increasing Cl_2 injection pressure and has the same slope as the upper curve obtained in the absence of the discharge. The shift in Cl_2 pressure due to the presence of the 5W microwave discharge reflects the absolute amount of Cl_2 which combined with the reactive species formed in C_2F_6 as it passes through the discharge. From the data obtained in Fig. 17 it can be determined that the partial pressure of Cl_2 utilized in combining with radicals produced in the C_2F_6 as it passed through the discharge was $\sim 0.6 \times 10^{-2}$ Torr. Since the only significant product involving chlorine detected downstream from the discharge was CF_3Cl a conclusion is reached that essentially all of the Cl_2 which reacted in the flow system resulted in the formation of CF_3Cl . Thus ~ 0.012 Torr of CF_3Cl formed in the flowing system. The relative number of CF_3Cl molecules obtained per unit of microwave energy, or relative efficiency, is plotted as a function of microwave power in Fig. 18 for the case where 0.02 Torr of C_2F_6 is flowing through the discharge at 100 cm/sec and where excess Cl_2 is injected at the orifice just below the discharge. It is seen from the figure that the relative efficiency increases sharply as the discharge power is reduced. Since the discharge could not be maintained for an absorbed power level at the microwave cavity of less than 4 W, it was not possible with the presently used microwave discharge system to examine lower power discharges. It appears that much, if not most of the microwave energy absorbed at the cavity is being dissipated in the flow tube rather than in the gas and it is expected that absolute CF_3 formation efficiencies will increase by improving the discharge system. An RF generator operating in the range of 10 MC is to be utilized in setting up a low energy gas discharge system. If more of the discharge energy can be coupled into the gas with this improved system, then significant improvements in the CF_3 generation efficiencies should be realized.

Initial experiments have been performed where the Cl_2 is pre-mixed with C_2F_6 before passing through the discharge. The relative CF_3Cl formation efficiency appears to be more than 3 times greater in this case than was observed for the case where Cl_2 is injected immediately below the discharge.

CF₃CL YIELD EFFICIENCY AS
A FUNCTION OF MICROWAVE
DISCHARGE POWER

The relative efficiency of CF_3Cl formation as a function of microwave cavity input power for a stream of C_2F_6 with downstream injection of Cl_2 .

Figure 18

The main reason for this increase in efficiency must likely be lower than that for the case where Cl_2 passes through the discharge alone with C_2F_6 . There is now favorable competition for CF_3Cl formation in the discharge itself as compared to CF_3 recombination to form C_2F_6 .

With a microwave discharge power dissipation of 4 W and using a flow rate of 100 cm/sec, the figure of merit τ for the formation of CF_3Cl in a $\text{C}_2\text{F}_6 + \text{Cl}_2$ mixture is approximately 1/2%. This value for τ is not considered at this time to be an optimum value since additional studies are to be carried out with the improved discharge system. A postscript value for τ for the formation of CF_3Cl using a 1.5 MHz r-f discharge is 4%. It further remains to be established what τ will be for the formation of CF_3I rather than CF_3Cl .

The results of this study have shown that CF_3 radicals can be produced in flowing C_2F_6 and that it is possible to react these radicals with Cl_2 to form CF_3Cl . The relative formation efficiency of CF_3Cl with respect to the formation of the undesirable compound C_2F_6 can be increased by working at low discharge power levels, low C_2F_6 pressures and relatively high flow rates. The CF_3Cl formation efficiency in the present laboratory setup is greatly improved by pre-mixing C_2F_6 with Cl_2 upstream from the discharge rather than injecting Cl_2 into the flow immediately downstream from the discharge.

2.6. Energy Cost Analysis of Cooling the Laser Head

Regardless of what pump source is used for producing the iodine atomic laser as much as 98% of the energy supplied to the pump will have to be carried off as waste heat. For a 100 kW arm in an iodine laser system $\sim 10^7$ J must be disposed of with some regard for the ecosystem in which the power plant is located. A recent survey³⁹ of the estimated resource requirements for alternative power plant cooling techniques considered the issue of water management from a water, land and energy standpoint. Table III summarizes some of these findings for an atomic iodine laser having an electrical figure of merit of 0.02 and with a ΔT of 11°C between the input and output cooling water.

The water requirements quoted in Table III are in gallons of water needed to produce a kilowatt hour of atomic iodine laser output. In the western United States the construction of new power plants has long been restricted based on the need for cooling water.

TABLE III

Estimated Resource Requirements for
Alternative Cooling Techniques

Cooling Method	Water		Energy (Figure of Merit)
	Thru-Put (Gal/kWh)	Consumption (Gal/kWh)	
Once-through	945	5.9	31.8
Cooling Ponds	945	5.9	28.2
Mechanical Draft Wet Towers	945	17.7	10.6
Mechanical Draft Dry Towers	-	-	2.65

The energy requirements quoted in Table III are given in terms of a figure of merit. The figure of merit is obtained by taking some fixed amount of energy produced by an atomic iodine laser and dividing that energy by the amount of energy needed to carry off 50 times that energy in waste heat. The smallest figure of merit is 2.65 for mechanical draft dry cooling towers. Comparing this with the largest figure of merit for pump sources, 0.09, we see that cooling of the laser head is an insignificant energy cost relative to the cost of pumping the laser.

2.7. Flow Cycle Efficiency

2.7.1 Description of Flow Cycle Loop

In order to consider the energy consumption in circulating, cooling, etc. the laser gas mixture, we have had to choose a specific design for the circulation loop. Figure 13 is a schematic representation of a closed loop flow cycle design upon which we have chosen to base our calculations. The equilibrated laser gas mixture consisting of Ar, RI, I_2 and R_2 enters the laser amplifier through a series of flow straighteners. At this point, a cushion of Ar (~ 10% of the amplifier cross section) is injected along the amplifier walls to keep products of the photolysis from depositing on the windows between the UV pump source and the laser gas. As the gas exits from the amplifier it passes through a set of acoustic dampers followed by cooling baffles upon which excess I_2 precipitates. The laser gas then proceeds to a point at which 10% of the gas is removed for cleanup and 90% continues around the loop. Processing only 10% of the gas allows a buildup of R_2 in the system, but the R_2 equilibrium value is not large enough to affect the laser performance. The 10% level was chosen to match the excess Ar added to the loop as a cushion. Also, since the bulk of the gas is Ar, processing only a small percentage of the mixture reduces the repetitious work on the Ar. This 10% removed from the stream is passed through a membrane or series of membranes upon which the I_2 , RI and R_2 collect. The clean Ar returns to be injected as the cushion gas, thus completing that loop. The R_2 , RI, and I_2 collected on the membranes are removed and separated. The RI is returned to the flow loop and the R_2 and I_2 are removed for chemical regeneration (treated earlier). The 90% of the flow which continued around the loop has the proper Ar concentration but is low on RI. At this

Conceptual flow cycle for a closed loop-iodine laser system.

Figure 19

point, RI flow rate is controlled, the chemical concentration facility is injected into the flow chamber at the proper concentration level. The mixture now must be thoroughly mixed because we not only have inhomogeneities following RI injection, but also only alternate slugs of gas have been exposed to flashlamp irradiation and thus to RI decomposition, R_2 formation, etc. Following the mix chamber, a pump system brings the pressure back to the level for injection into the lower end.

3.3.3. Development of Equations for Amplifier Section

We now turn to the development of amplifier equations. This shows us the level of output and the relative value of parameters of the system to the different processes. These equations will now be developed and then an energy consumption study of the energy consumption in the various stages of the flow will be outlined above.

The window damage limit $(E/F)_{\max}$ establishes the quantity N_I^*L by the relationship;

$$N_I^*L = F_{\max} \frac{b}{h\nu} \text{ (cm}^{-2}\text{)} \quad (15)$$

where N_I^* is the number density of excited iodine atoms, L is the amplifier length, b is the degeneracy ratio, h is Planck's constant and ν is the laser frequency. The window damage limit and the required output energy E establish the aperture size H

$$H = (E/F_{\max})^{1/2} \text{ (cm)} \quad (16)$$

where E is the output energy obtained when 90% of the extractable energy is removed from the amplifier. The requirement of uniform pumping establishes the number density of parent molecules N_{RI}

$$N_{RI} = \frac{1.6}{\tau_{uv} H} \text{ (cm}^{-3}\text{)} \quad (17)$$

where τ_{uv} is the ultraviolet absorption cross section of the parent and the factor 1.6 gives pumping uniformity of $\pm 17\%$ using a Beer's law formulation and the absence of any strong bleaching.

If we assume 25% photolysis of the RI which is common for current iodine systems, we have established N_I^* since

$$N_I^* = 0.25 N_{RI} = \frac{0.4}{\sigma_{uv} H} \text{ (cm}^{-3}\text{)} \quad (18)$$

L can now be determined from (15). Broadening of the RI absorption spectrum is not included explicitly in the equation developed here. Note, however, that broadening effectively reduces σ_{uv} , hence increasing N_I^* and reducing L. Next, the requirement that the amplifier not self-oscillate establishes the laser cross section σ and hence the buffer gas pressure P_B .

$$\sigma N_I^* L = \ln G_{\max} = \mathcal{G} \quad (19)$$

where $\mathcal{G}$ is the maximum gain in Nepers before self-oscillation (a typical value for an atomic iodine laser is $\mathcal{G} \approx 8$) and

$$\sigma = \frac{\lambda^2 A}{4\pi^2 B P_B} \quad (\text{cm}^2) \quad (20)$$

where λ is the laser wavelength, A is the Einstein coefficient, and B is the line-broadening coefficient in MHz/Torr. Evaluating known quantities, the buffer gas pressure is given by

$$P_B = \frac{3.45 \times 10^4 F_{\max}}{B \mathcal{G}} \quad (\text{Torr}) \quad (21)$$

Other quantities of interest are

$$L = 9.93 (F_{\max} E)^{1/2} \quad (\text{cm}) \quad (22a)$$

$$H = (E/F_{\max})^{1/2} \quad (\text{cm}) \quad (22b)$$

$$N_{RI} = 4 \times 10^{18} (F_{\max}/E)^{1/2} \quad (\text{cm}^{-3}) \quad (22c)$$

It should be noted that the flux on the output window is crucial in determining the size of the amplifier for a given output energy. It should be possible to maintain adequate medium homogeneity over the maximum length considered in this report, approximately 50 meters.

2.7.2.2. Bleaching the Laser Medium

To maximally utilize the laser working gas and at the same time avoid severe pumping nonuniformities it would be advantageous to bleach the laser medium. For a photolytic source of sufficient intensity, the dissociating transition that produces I^* from the parent molecule can be saturated. The lower limit to the required energy fluence is $E_s = c_p/\sigma_{uv} \text{ (J/cm}^2\text{)}$, where c_p

is the pump photon energy. For C_3F_7I dissociation by KrF laser radiation at 249 nm, $E_s \approx 2 \text{ J/cm}^2$. (Stokes shifting of the KrF laser radiation to 277 nm by H_2 would lower E_s to about 1 J/cm^2 .) Under bleaching conditions, the size of the I^* amplifier is determined by the available pump fluence E_p . For the three conditions of two-sided irradiation, $E_p \gg E_s$, and efficient use of pump photons, E_p satisfies:

$$E_p = N_{RI} \left(\frac{h}{2} \right) \epsilon_p = N_I^* \left(\frac{h}{2} \right) \epsilon_p \text{ joules/cm}^2 \quad (23)$$

Equation 15 can be rewritten to give:

$$L = \frac{F_{\max}}{N_I^*} \left(\frac{b}{h\nu} \right) \quad (24)$$

This equation can be used in conjunction with Eq. 23 to evaluate the length for a particular pump fluence, i.e., to relate N_I^* to E_p .

The upper limit to N_I^* in the present case is dictated by the deactivation process

which has a rate coefficient of $2 \times 10^{-13} \text{ cm}^3 \cdot \text{sec}^{-1}$. An I^* storage time of 10 μsec restricts the initial I^* density to $N_I^* \leq 5 \times 10^{17} \text{ cm}^{-3}$. With this limitation one has from Eq. 24:

$$L \geq 20 \frac{F_{\max}}{\text{cm}} \quad (25)$$

Equation 25 is to be compared with Eq. 22 (a). The difference between these arises because of the assumptions concerning N_I^* . For high energies of interest, Eq. 25 predicts a much shorter length. Hence, with complete bleaching of the laser medium a short high energy amplifier is possible.

2.7.3. Development of Equations for Flow Energy Consumption

Now let us add flow to the problem. If we assume that the flow ducts have a cross sectional area LH throughout, the linear flow rate U is uniform and equal to

$$U = \frac{HR}{D} = \frac{R}{D} (E/F_{\max})^{\frac{1}{2}} \text{ (cm/sec)} \quad (26)$$

where R is the pulse repetition rate and D is the duty factor of the flow. The volumetric flow rate V is given by

$$V = 1.1 \text{ LHU} = 1.1 V \frac{R}{D} = 1.09 \frac{R}{D} \frac{E^{3/2}}{F_{\max}^{1/2}} \text{ (cm}^3/\text{sec)} \quad (27)$$

where V is the volume pumped and the factor 1.1 accounts for the buffer gas cushion.

It will now be useful to calculate the mass flow rate. Let's assume the mixture is pure buffer gas. Then the density of the gas ρ is given by

$$\rho \cong \rho_B = \frac{P_B M_B}{R T} = 5.38 \times 10^{-8} P_B M_B \text{ (g/cm}^3\text{)} \quad (28)$$

at 298° K and where M_B is the molecular weight of the buffer gas or in terms of our other parameters

$$\rho_B = 1.86 \times 10^{-3} \frac{M_B F_{\max}}{B \phi} \text{ (g/cm}^3\text{)} . \quad (29)$$

The mass flow rate is given by

$$\dot{m} = \rho V = 2.02 \times 10^{-3} \frac{M_B R E^{3/2}}{B \phi} F_{\max}^{1/2} \text{ (g/sec)} . \quad (30)$$

Now let us turn to the development of the equations necessary to calculate the energy consumed to circulate the laser gas mixture around the flow loop. Let ϕ be the power required to drive the compressor circulating the gas around the loop

$$\phi = \frac{P_o U A}{\eta_c} \left(\frac{\gamma}{\gamma-1} \right) [\delta^{(\gamma-1)/\gamma} - 1], \text{ (watts)} \quad (31)$$

where P_o is the compressor output pressure, A is the equivalent duct area ($L \times E$), η_c is the compressor efficiency, $\gamma = C_p/C_v$, and $\delta = P_o/(P_o - \Delta P)$ with ΔP equal to the loop pressure drop. For our condition $P_o = P_3$, $\gamma = 5/3$ and if ΔP is small enough (to be shown below) that

$$\begin{aligned} \delta^{(\gamma-1)/\gamma} - 1 &= \left[P_o/(P_o - \Delta P) \right]^{0.4} - 1 = \\ (1 + \frac{\Delta P}{P_o})^{0.4} - 1 &= 0.4 \frac{\Delta P}{P_o} \end{aligned} \quad (32)$$

then

$$\phi = \frac{U L H}{\eta_c} \Delta P = \frac{1.32 \times 10^{-3} E^{3/2}}{\eta_{DF} \max} R \Delta P \text{ (watts)} \quad (33)$$

We now need to estimate the pressure drop for the various elements in the flow cycle loop. For any element in the loop, the pressure drop is given by

$$\Delta P = 3.75 \times 10^{-4} \rho U^2 \frac{fL}{d}, \quad (\text{Torr}) \quad (34)$$

where ρ is the density of gas in gm/cm^3 , L/d is the effective length/diameter ratio of the element and f is the friction factor which depends on the Reynold number and wall roughness. (For our flow regime, $f = 0.01$ for very smooth walls and $f = 0.04$ for moderately rough walls.) In terms of the earlier parameters,

$$\Delta P = 6.98 \times 10^{-7} \frac{M_B R^2 E}{B D^2} \frac{fL}{d} \quad (35)$$

for a single element in the loop. Summing all elements we then find the power consumption from

$$\phi = 9.21 \times 10^{-10} \frac{E^{5/2} R^2 M_B}{\eta_B D^3 F_{\max}} \Sigma \left(\frac{fL}{d} \right) \quad (36)$$

Now the figure of merit of the circulation system will be

$$\eta_{\text{CIRC}} = \frac{1.09 \times 10^9 \eta_B D^3 F_{\max}^{1/2}}{E^{3/2} R^2 M_B \Sigma \left(\frac{fL}{d} \right)} \quad (37)$$

2.7.4. Evaluation of Gas Circulation Figure of Merit

Let us now evaluate the figure of merit for various values of the parameters. We will fix the following parameters. The pulse repetition rate R at 3, the flow duty cycle D at 0.5 the window damage limit $F_{\max}$ at 10 J/cm^2 , the maximum gain B at 8, and the compressor efficiency η at 0.85. Thus we have

$$\eta_{\text{CIRC}} = \frac{3.25 \times 10^8 B}{E^{3/2} \Sigma \left(\frac{fL}{d} \right)} \quad (38)$$

Since M_B fixes B , this leaves us with 3 parameters for which to calculate the figure of merit for gas circulation.

Figure 20 presents a plot of η_{CIRC} vs $\Sigma \left(\frac{fL}{d} \right)$ for an assumed output energy of 30 KJ. The two curves represent the use of Ar and He as the buffer gas. Best estimates for our flow cycle loop put $\Sigma \left(\frac{fL}{d} \right)$ at ~ 9 .

$\eta_{circ} \text{ vs. } \Sigma(I/A) \text{ for an assumed laser output energy of } 30 \text{ kJ.}$

FIGURE 20

Figure 21

Figure 21 presents a plot of η_{CIRC} vs. E for an assumed $\Sigma(\frac{f_i}{d}) = 10$. Again the two curves represent the use of either He or Ar as the buffer gas. In examination of the curves, the scale difference for η_{CIRC} calculated with Ar or He should be noted.

The first conclusion drawn from these graphs is that He is the choice as buffer gas. The use of He does raise the total pressure of the system since the pressure broadening coefficient is smaller than for Ar. However, even using Ar as the buffer gas, the figure of merit is large enough not to be the limiting quantity in the overall system efficiency. We can conclude that for an amplifier of 30 kJ output with He as the buffer gas and an estimated $\Sigma(\frac{f_i}{d}) = 10$, the figure of merit for circulating the gas would be ~ 5 .

2.7.5. Evaluation of Figure of Merit for Other Elements of Flow Loop

We will now estimate the energy consumption and figure of merit for heat removal from the laser gas. If we assume that the total chemistry of the system is thermoneutral (a valid assumption in systems where loss to R_2 and I_2 formation is small), then for a 4.5 eV pump photon (275 nm), a 1 eV laser photon and 60% extraction, we have ~ 4 eV of energy per excited iodine atom created which will be lost to the gas as heat and must be removed. Since the laser energy out is 0.6 eV on the average, we have a relationship that the waste heat is 6.6 E. This waste heat will create a temperature rise of $\sim 1^\circ \text{C}$ in the gas. Let us assume that the refrigeration cycle heat exchanger operates at $T = 10^\circ$, where T is the ambient room temperature. The thermodynamic efficiency of such a device is given by

$$\frac{q_c}{w} = \frac{T_G}{(T + 10) - T_G} \quad (39)$$

where q_c is the heat removed, w is the work required and T_G is the temperature of the laser gas. Because of the small temperature rise in the gas, let us assume that $T_G = T + 1 \cong T$. This yields

$$\frac{q_c}{w} = \frac{T}{9} \quad (40)$$

The overall efficiency will be the thermodynamic efficiency times the mechanical efficiency of the refrigeration process η_R . Now we have

$$w = \frac{9}{\eta_R T} \cdot q_c = \frac{9}{\eta_R T} (6.6 \text{ E}) \quad (41)$$

The figure of merit for gas cooling η_{gc} will be given by

$$\eta_{gc} = \frac{E}{W} = \frac{1}{R(T/6.0)} \quad (4.2)$$

If we assume that the ambient temperature is 300°K and that the mechanical efficiency of the refrigerator is .5 then the gas cooling figure of merit is 2.9.

It is somewhat difficult to estimate accurately the energy consumption in the buffer gas clean-up element of the flow loop. However, the primary energy cost will be in pumping the Ar through the membrane or across a pressure drop. It is our feeling that the membrane can be of sufficient size that the pressure drop across it will be of the order of the pressure drop in circulating the gas around the flow loop cycle. However, because of the 10% processing at this stage, even if the pressure drop were 10 times larger than for the circulation loop, the figure of merit would be 5. For an assumed considerable pressure drop, the figure of merit would be 50.

The final step for which we must calculate a figure of merit is the cryogenic separation of RI, I₂ and R₂. From the equations presented above, we can calculate the mass flow rate of RI in the system as a function of laser energy, repetition rate and flow duty cycle. We have assumed above that R = 1 and D = 0.5. Since the mass flow rate varies linearly with laser energy we will base our calculations on a laser energy of 30 kJ with the knowledge of its linear dependence on E. The mass flow rate of RI will be 1.3 moles/sec at a laser energy of 30 kJ. The mass flow of R₂ will be 2.5 times that of RI for the worst case parent molecule or 3.3 moles/sec. I₂ will make a negligible contribution to this calculation because it has already been mostly removed from the stream. We process 10% of this flow or .13 moles/sec of RI and .33 moles/sec of R₂. We shall assume that we cool both to liquid nitrogen temperature. (In reality we would cool each to a different temperature probably somewhat higher than liquid nitrogen.) We shall also assume a constant heat capacity of 17 cal/deg. mole for RI and 25 cal/deg. mole for R₂. While for these molecules the heat capacity would decrease with temperature as the vibrational modes froze out, we will make the assumption of constant heat capacity and neglect the heat of condensation. Using the above numbers and a mechanical refrigerator efficiency of 0.5, we obtain a value of ~ 16 kJ/sec of energy required to cool the RI and R₂

in the 10% split off of the flow to 77° K. This is compared to a laser energy of 90 kJ/sec or a figure of merit of 6 for this stage.

2.7.6 Conclusion

The total flow cycle figure of merit η_T is given by

$$\frac{1}{\eta_T} = \frac{1}{\eta_{\text{CIRC}}} + \frac{1}{\eta_{\text{gc}}} + \frac{1}{\eta_S} + \frac{1}{\eta_C} \quad (44)$$

where η_S is the figure of merit for buffer gas cleanup and η_C is the figure of merit for the cryogenic cooling of laser gas byproducts. Thus, for a laser of 30 kJ output energy, 3 Hz, 50% duty cycle, He buffer gas, and other parameters as specified earlier, we have a total flow cycle figure of merit equal to ~ 1 . This number should be compared to projected figures of merit in the range of 0.01 to 0.03 for the pumping mechanism. This value will go down for higher laser output energies or it will go down for use of Ar as a buffer gas. However, it appears that even allowing for some large errors, the total flow cycle efficiency will not limit the overall system efficiency.

CHAPTER 3: Work Required to Complete the Development of the Iodine Laser System

3.1 Introduction

As has been demonstrated in Chapter 2, the atomic iodine laser system is physically well understood. Generation of atomic iodine laser pulses having the qualities required for their use in the inertial confinement fusion program can be accomplished. The only barriers to the successful utilization of the atomic iodine laser in this program are ones of efficiency and reliability. It is shown in Chapter 2 that these barriers are associated primarily with the photolytic pumping and chemical regeneration steps. This chapter indicates briefly the nature of the work which must be performed in order to determine the optimum figures of merit of these two steps and the projected reliability of the pump source. A brief discussion of a pulsed atomic iodine laser based on the chemical generation of excited iodine atoms is also given.

3.2 Pump Sources

The development of a suitable pump source remains the major obstacle to the use of the atomic iodine laser in the ICF program. As can be seen in Chapter 2, no pump source in its present state of technology allows the design of an atomic iodine laser system with $>1\%$ overall efficiency. However, several prospects cannot be ruled out as potential candidates for an acceptable pump.

Metal-doped Xe flashlamps and surface discharges have shown promise in initial studies. Both tend to enhance the UV emission by increasing the plasma density in the discharge, and future studies should be performed to see whether either can achieve the black-body limit of $\approx 15\%$ fluorescence efficiency in the atomic iodine laser pumpband. If the black-body emission is realized and if the energy efficiency of other aspects of the overall system, e.g. chemical recovery, proves to be favorable, then an overall system efficiency of 1% should be attainable. Of course, these two pump sources also require a great deal of technological development in order to achieve a suitable lifetime.

Both metal-doped flashlamps and surface discharges are broadband emitters. If either is used in a large atomic iodine laser, overall system

efficiency would be greatly improved and system length appreciably reduced through a broadening of the parent molecule UV absorption band. As detailed in Chapter 2, broadening by greater than a factor of 2 is accompanied by an unacceptable degree of decomposition in CF_3I , but other molecules have not been investigated. The potential benefits of broadening are great enough that a thorough experimental study of molecules such as $\text{C}_2\text{F}_4\text{I}$ should be made. Also, smaller than a factor of 2 broadening may not be accompanied by an unacceptable degree of decomposition and small improvements in efficiency may be sufficient to push this laser past the acceptable efficiency barrier.

The HgXe system has the potential of becoming an extremely efficient pump source. Unfortunately, present experimental evidence does not demonstrate this high efficiency. Experiments have been performed under non-optimal conditions, however, and it is important that experiments be performed under the conditions for which high efficiency is projected. If HgXe can indeed achieve a 40% fluorescence efficiency with an acceptable intensity, then the atomic iodine laser has a good chance of becoming an extremely attractive driver for an ICF reactor.

Low pressure Hg lamps have already been shown to have an acceptable fluorescence efficiency, but several experiments remain to be performed in order to determine whether they are applicable to the atomic iodine laser. First the chemical regeneration scheme must be shown to be suitably efficient, even if the pump pulsewidth is several msec. Second, the optical properties of a beam propagated through a long gain medium that has been pumped for several msec must be shown to be such that the beam is still focusable. Present projections suggest that both of the problems will be severe, but experiments should be performed to see whether this is in fact the case. These same comments apply to a long-pulse-width free-electron laser photolysis source.

It has been shown in Chapter 2 that the length of an atomic iodine laser amplifier can be greatly reduced if the photolytic pumping source has sufficient intensity to bleach the medium. Bleaching will probably be feasible only for narrow band pump sources. The source showing the most promise is KrF . Even though the projected electrical figure of merit for a laser pumped with KrF is presently < 0.01 , it may be useful to study the problem of scaling KrF systems to sufficient intensity ($\sim 5 \text{ J/cm}^2$) that they might be used to bleach an iodine laser medium.

3.3 Chemical regeneration

The development of a complete regeneration process is still in its early stages. Both the laser irradiation and the AC electric discharge schemes continue to be attractive as indicated in Chapter 2; both must be carried to a more advanced state of development before a choice can be made between them.

As indicated in Chapter 2, the regeneration figure of merit that can be attained experimentally using the laser irradiation scheme is consistent with the theory reported in reference 3, and with an overall laser efficiency of greater than 1%. It must now be determined that acceptable figures of merit are obtained when R_2 molecules other than C_2F_6 are used and when I_2 is substituted for H_2 . Presently the weakest aspect of the laser scheme is that it produces some undesired by-product. This product can be recycled fairly easily, but the extra processing required will reduce the regeneration figure of merit somewhat. An important goal of future work must be to determine whether by-products exist when R_2 , I_2 systems are studied and, if so, how to minimize the generation rate of by-products.

The AC electric discharge technique for chemical regeneration has been found to produce negligibly small amounts of by-products in R_2 , Cl_2 systems. It has recently been found that using MHz rather than GHz discharge frequencies enhances the efficiency of the technique, and the present regeneration figure of merit for the production of CF_3Cl from C_2F_6 and Cl_2 is 0.01. If similarly good selectivity and efficiency are found for R_2 , I_2 systems, the discharge technique will be more attractive than the laser technique.

3.4 Iodine Laser Based on Chemical Processes

Chapter 2 discusses a recently demonstrated, purely chemical technique for producing iodine laser action. The discussion in Chapter 2 focuses on questions of efficiency and concludes that an overall system efficiency of 6% may be attainable. For this reason alone, the chemically-driven iodine laser system should be investigated seriously. Air Force Weapons Laboratory personnel are involved with investigating CW applications of the system, but no detailed investigation of its application to the efficient production of

short pulses is presently under way. A pulsed iodine laser utilizing chemical techniques would be of much different design than one using photolytic pumping for several reasons. For instance, the total operating pressure would have to be low to avoid iodine recombination. The stimulated emission cross section could not therefore be controlled by the addition of buffer gas. Inhomogeneous broadening using a non-uniform magnetic field might be used in the place of the buffer gas, but the amplifier input signal would then have to be very broad spectrally. The point to be considered is that pursuing a chemically-driven iodine laser represents a major change in direction. This change may well be worth making when two points in addition to efficiency are considered. First, a chemically-driven iodine laser avoids the use of pulsed power systems, thus greatly enhancing system reliability. Second, the chemical-production system is quasi-CW and lends itself very easily to the design of lasers with high repetition rate.

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