# **Design and optimization of a pulsed CO<sub>2</sub> laser for laser ultrasonic applications**

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Laser ultrasonics is currently the optimal method for non-destructive testing of composite materials in the aerospace industry. The process is based on a laser-generated, ultrasound wave which propagates inside the composite. The response at the material surface is detected and converted into a defect map across the aircraft. The design and optimization of a laser system for this application, together with the basic science involved, is reviewed in this paper. This includes the optimization of laser parameters, such as output couplers and gas mixture, and the impact these choices have on the laser chemistry. We present a theory for the catalytic recombination of the gas which shows excellent agreement with experiment. Finally, an operating laser system for this application, yielding a sixfold improvement in performance over conventional laser systems, is described.

#### Introduction

Polymer–matrix composites are increasingly used in the aerospace industry, particularly in the manufacture of modern fighter planes. <sup>1-3</sup> The number and complexity of such composites are also increasing steadily. The aerospace industry requires non-destructive testing (NDT) of all parts during the manufacturing process. In the case of composite materials, testing is particularly necessary for detecting the presence of delaminations and inclusions. The conventional approach involves the use of ultrasonics, based on water jets and piezoelectric transducers. A requirement for this process is that the transducers must be normal to the surface to ensure good signal-to-noise ratio. Since most modern aircraft parts have a complex geometry, this process is slow and therefore expensive.

An alternative ultrasonic inspection technique is laser ultrasonics (LU). In this technique two lasers are used to illuminate the part: a short pulse laser with a wavelength chosen so as to be absorbed by the material, and a second laser beam to detect the impact of the ultrasonic waves. The first laser pulse is rapidly absorbed by the material, causing fast thermal expansion, which results in ultrasonic waves. The wave amplitudes themselves are small enough not to cause any damage. The waves so generated propa-

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gate through the material and are reflected from interfaces (such as defects) inside the material. On returning to the surface, the reflected waves generate very small surface displacements of the material. A second, narrow bandwidth, frequency stable laser is used in conjunction with an interferometer to detect these mechanical displacements on the surface of the material. The advantage of LU is that these measurements can be done at large distances from the sources and at large angles with respect to the sample surface. This gives LU a competitive advantage and ensures that it is currently the NDT method of choice for the testing of composite materials in the aerospace industry.

For efficient and reliable operation, LU requires the efficient generation of ultrasound by the first laser pulse, which places stringent requirements on the laser system. The properties required of the laser pulse include good beam quality, a pulse of short duration (<100 ns), as well as a minimum energy (>200 mJ) and pulse repetition rate (>200 Hz) for the process to be suitable for industrial applications. Furthermore, since these systems will typically be used in a production environment, the final design must result in a laser system that is reliable (with low downtime) and economical. Numerical simulations and experimental investigations have indicated that generation efficiency can be improved by using short pulses in the 3–4  $\mu$ m and 10  $\mu$ m spectral regions. Short-pulse 10-μm radiation can be produced by transversely excited, atmospheric CO<sub>2</sub> (TEA CO<sub>2</sub>) lasers. Owing to the technological maturity of these lasers, they are particularly suited to industrial environments.

In this paper we discuss the design of a TEA  $\rm CO_2$  laser for LU applications. We cover the basic laser parameters to be optimized, and report experimental data for the optimization of the output coupler reflectivity and gas mix. The impact on laser chemistry is discussed in detail.

#### Laser parameters

It is useful to start with a summary of the important laser parameters for LU, and their interrelationships. Since the parameters are not independent of one another, the critical aspect of the parameter testing is to explore regimes in terms of combinations of parameters. Only in some cases can the parameters be chosen independently. For LU a strong ultrasonic signal at as high a repetition rate as possible must be generated. The strong signal ensures a good signal-to-noise ratio, while a higher repetition rate allows more samples to be tested in a given time, thereby increasing productivity. The LU signal is a product of the efficiency of the laser pulse in generating ultrasound, determined

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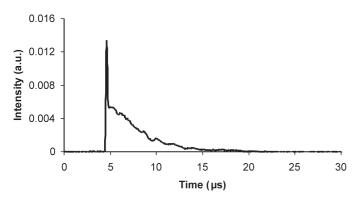


Fig. 1. A typical time pulse from a TEA CO<sub>2</sub> laser.

by wavelength and temporal characteristics, and the energy of the laser pulse determined by gain/loss effects inside the resonator.

A typical time pulse from a TEA  $\rm CO_2$  laser is shown in Fig. 1. It comprises a short duration 'spike' followed by a somewhat longer duration 'tail'. Since the total energy contained in the pulse is the time-integrated signal, a long tail implies that most of the energy is contained in the latter part of the pulse. The time profile of the laser pulse is of paramount importance for efficient LU which requires a very short time pulse (<100 ns), with as much of the energy in the spike as possible. Factors influencing the laser energy and time profile include gas mix, output coupler reflectivity (the amount of energy retained in the laser cavity), discharge volume and operating voltage amongst others.

Inevitably there are trade-offs. For example, high pulse energy from TEA  $CO_2$  lasers is usually associated with high  $N_2$  content in the laser gas mix; but high N<sub>2</sub> content is also associated with long time pulses. In general, the time envelope of the laser pulse is determined by the gain of the laser system. This in turn is largely influenced by both the reflectivity values of the cavity optics, and the gas mixture used in the laser. The gas mixture also has a direct influence (that is, not via the gain) on the time profile through the fraction of the excitation energy stored in the excited  $N_2$  upper energy levels. This energy is transferred to the upper energy levels of the CO<sub>2</sub> molecule through collisions, resulting in a delayed energy output from the laser in the form of a characteristic 'tail' observed in many pulses. In Fig. 1, for example, the tail lasts for roughly 15  $\mu$ s. Thus high  $N_2$  concentrations are usually associated with high energy and prolonged pulses with long tails. The general trend in the course of optimization for shorter pulses, was towards higher CO<sub>2</sub> fractions with lower N<sub>2</sub> fractions. As the pulse time envelope and the tail are reduced, the total energy output drops. The energy drop was minimized through careful selection of the operating voltage (since the energy deposited into the laser discharge is proportional to the square of the operating voltage), and output coupler reflectivity. The problem was to find a working parameter set that maximized the LU signal, without adversely affecting the laser stability.

# **Output coupler**

There is an optimal output coupler (OC) reflectivity for a given laser resonator in terms of energy extraction from the cavity. The graph in Fig. 2A illustrates this by plotting the efficiency of energy extraction from the laser as a function of the OC reflectivity. In this case, the optimal OC is roughly R = 0.55 (or 55%). The choice of OC also impacts on the amount of energy circulating inside the laser cavity, which affects the lifetime of the cavity optics. The ratio of the energy inside the cavity to the energy outside is termed *intra-cavity enhancement*. High enhancement values usually indicate the likelihood of optical damage, since the optical energy density on each optic inside the cavity is high.

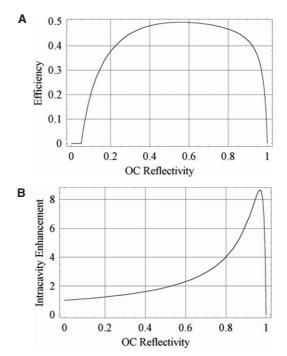


Fig. 2. A, An example of the dependence of the energy extracted from the laser cavity as a function of the output coupler (OC) reflectivity. The graph is calculated for a low-gain resonator, and changes with laser gain and gain length. [The optimal OC, where the maximum output energy occurs, has a reflectivity near 0.55 (or 55%).] B, Intra-cavity enhancement as a function of the output coupler reflectivity.

The dependence of intra-cavity enhancement on output coupler reflectivity is shown in Fig. 2B. Clearly the enhancement factor rapidly increases as the OC reflectivity nears  $R=0.95\ (R=1$  implies the maximum 100% is reflected). This graph is useful only once the output energy is known as a function of OC reflectivity, since only then can the circulating energy inside the cavity be calculated and compared to known optical damage values. Clearly it is desirable to have the circulating energy well below the optical damage point.

Four output couplers were used in experiments, with percentage reflectivities of 36%, 50%, 55% and 70%. In addition to measuring the output energy as a function of OC, the LU signal was measured and the efficiency of the pulse in generating the signal was computed as a V/J value; that is, the amount of signal in volts generated per joule of input energy. These experiments were repeated for a range of gas mixtures (discussed in detail in the next section) in order to find the balance between high energy extraction and good LU efficiency, equivalent to short time pulse generation. Figure 3 shows a parametric plot of the LU efficiency (units of V/J) and measured output energies for the four tested OCs, with various high-voltage (HV) settings (essentially various gain conditions) and gas mixtures. It is evident that, in general, lower OC reflectivity values give better LU efficiency at lower energies, indicating that such pulses have 'better' pulse time characteristics for this application. As expected, a comparison of the pulse characteristics reveals that short pulses with small tails result in a better LU signal. The spread in results for a given OC is due to the gas mix and HV ranges used. Since the spread is in both efficiency and energy, one can deduce that these parameters also influence the time pulse characteristics.

In summary, if all other parameters are kept constant, then increasing the output coupler reflectivity has a detrimental effect on the acoustic efficiency. The broad trend is for lower reflectivity OCs to outperform higher reflectivity OCs in terms of LU efficiency, while the reverse is true in terms of energy extraction from the laser. Ideally, therefore, one would like to use

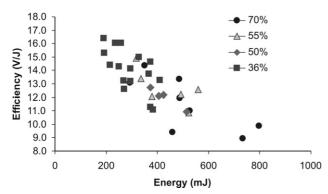


Fig. 3. The LU signal efficiency, as a function of the laser energy for various combinations of OC reflectivities, gas mix and laser operating voltage.

as low an output coupler reflectivity as is possible, and compensate for the energy loss in other ways, such as by adjusting the HV or optimizing the gas mix used.

#### Gas miyes

Traditionally, a high energy TEA  $CO_2$  laser (with a closed gas system) has a He content of roughly 80% or more and a  $CO_2$  content of less than a few per cent. The remainder would be mostly  $N_2$ , with sometimes some  $H_2$  and CO to allow the gas to be 'self-catalytic' (the  $H_2$  acts as a catalyst in the recombination of CO and  $O_2$  to form  $CO_2$ ). A 20%  $CO_2$  fraction would be considered a very 'rich' mix. Such traditional mixes result in pulse characteristics similar to that shown in Fig. 1 — undesirable for LU applications.

Following the results shown in Fig. 3, various gas mixtures were tested for energy extraction and time pulse characteristics, each with a different He and CO<sub>2</sub> molecular fraction. Most tests involved mixes with less than or equal to 50% He and high CO<sub>2</sub> fractions. The general trend observed was towards shorter time pulses using higher CO<sub>2</sub> fractions and lower N<sub>2</sub> fractions. A pulse with excellent time characteristics for LU is shown in Fig. 4. The pulse was measured using a gas with a 56% CO<sub>2</sub> fraction by volume. The resulting pulse had a full width, half-maximum (FWHM) time duration of 51 ns, with no discernible tail and with more than 85% of the laser energy present in the first 100 ns. Such a pulse shows a greater than 2-fold improvement in LU signal when compared with previously quoted values. However, lowering the N<sub>2</sub> fraction decreased the total energy in the pulse. Increasing the HV compensated for the lower energy, but this in turn placed strain on the electrical components: the potential difference across the electrodes can increase to as much as 40 kV, placing strain on all electrical and insulating components.

Another problem with such radical changes in the gas mix is the impact the change in density has on the refractive index of the medium through the Gladstone–Dale law,  $n-1=k\rho$ , where n is the refractive index and  $\rho$  the density. This was not investigated in this study, but the density of the medium has been observed to play an important role in discharge instabilities, such as acoustic waves, which are generally not a problem in equivalent systems that have much higher He fractions. Thus, the gas mix cannot be chosen to optimize the pulse without considering the consequence of this choice on the rest of the system. Another problem observed with the use of high  $CO_2$  and low He fractions, arose from the chemistry of the laser in the course of extended operation. This is covered in the next section.

# Laser chemistry

The CO<sub>2</sub> gas fraction in a TEA CO<sub>2</sub> gas laser decomposes over

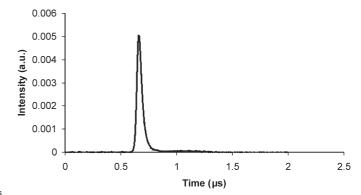


Fig. 4. The time pulse (51 ns) measured from the laser with a  $CO_2$  content of 56%, and a 36% OC reflectivity. (More than 85% of the pulse energy is in the first 100 ns.)

time to mainly CO and O2 and a small amount of other by-products, as a result of the electrical discharge between two electrodes:  $2CO_2 \rightarrow 2CO + O_2$ . Thus, in the absence of other reactions, the CO<sub>2</sub> concentration, [CO<sub>2</sub>], decreases with time while the  $O_2$  and CO concentrations ( $[O_2]$  and [CO], respectively) increase with time. This is a problem for a number of reasons. First, as [CO<sub>2</sub>] decreases so the power output from the laser decreases. Secondly, as  $[O_2]$  increases so the laser discharge becomes very unstable, with highly variable pulse-to-pulse energy values. Figure 5 shows a plot of laser pulse energy (total energy contained in each pulse) with oxygen concentration measured at simultaneous times (400 Hz measurements, that is, at the repetition rate of the laser). We have verified (not reported here) that the energy decrease is due to the decrease in [CO<sub>2</sub>]. Additionally, there is a marked increase in the pulse-to-pulse energy variability at high oxygen concentration levels. We attribute this to the adverse effects of O<sub>2</sub> in such electrical discharges, which often lead to electrical arcing. Experiments over more than a billion pulses suggest that the relationship between energy and O<sub>2</sub> content is also affected by the presence of water vapour (unreported data). Further experiments suggest that the increase in [CO] does not have any deleterious effects on the laser power output. Thus, the decomposition of CO<sub>2</sub> contributes both to a decrease in laser energy and an increase in laser instability. Needless to say, both these effects need to be

This decomposition process can be reduced, or its effects negated, by the use of self-catalytic mixes to regenerate the  $CO_2$ , or by employing a blow-through system which continuously replenishes the gas. As already discussed, to achieve the laser specifications for LU, very rich mixes with at least a 40%  $CO_2$  and low nitrogen content, are required. Self-catalytic mixes simply cannot reconvert the resulting  $O_2$  and CO at the required rate to

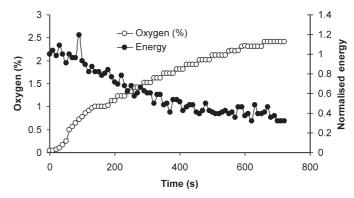


Fig. 5. Simultaneous measurements of the pulse energy versus oxygen levels in the laser at  $400\ \text{Hz}.$ 

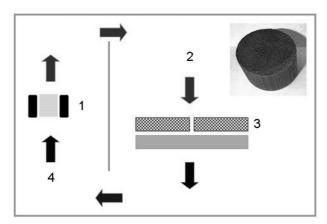


Fig. 6. Schematic representation of the gas system. A photograph of the catalysts used is shown in the inset.

meet these requirements and instead find use in low power CO<sub>2</sub> lasers with only a few per cent CO<sub>2</sub>. As an alternative, the laser was operated with internal, room temperature catalysts, mounted downstream of the gas flow through the discharge volume but just upstream of the heat exchanger. Thus the hot depleted gas first passed through the catalysts for regeneration, and then through the heat exchanger for cooling. In an ideal system, a single round trip would regenerate the initial gas conditions, a cool gas still rich in CO<sub>2</sub>, so that the process repeats endlessly. The role of the catalysts is to approximate this state by reversing the breakdown reaction through the oxidation of CO back to CO<sub>2</sub>. In the case of room temperature catalysts (usually Pt/Sn based), the mechanism is not well understood. Some studies<sup>4</sup> indicate that excess CO improves catalyst performance, as well as reducing O<sub>2</sub> formation in the discharge, while other studies<sup>5,6</sup> show the converse. It seems generally agreed<sup>4-7</sup> that excess  $O_2$  is undesirable for both laser action and catalyst activity. In the case of a Sn–Pt/SiO<sub>2</sub> catalyst, the presence of pure O<sub>2</sub> for a period of 15 minutes resulted in complete loss of catalyst activity.<sup>5</sup>

# Model

In this section we derive an expression that describes the time change in [O<sub>2</sub>] in a laser when catalysts are present and when absent. A schematic of the system modelled is shown in Fig. 6. The laser is modelled as a closed gas system with a channelled gas flow in the clockwise direction. At stage 1, [O<sub>2</sub>] increases due to dissociation of the CO<sub>2</sub> molecules. The gas increases in temperature (see Equation (7) later) and, through collisions, facilitates a small re-conversion of the  $O_2$  and CO back to  $CO_2$ which we depict as stage 2. This gas then passes through the catalysts (stage 3), which we assume will preferentially drives the reverse reaction back to CO<sub>2</sub> (since the reaction is not operating at equilibrium). The gas then passes through a system of heat exchangers to remove any excess heat, and return the cool gas to the discharge to repeat the cycle, starting again at position 4. The system has been modelled as a continuous flow system but with the gas flow continuously re-entered into the system. The three stages to the process give rise to coupled differential equations, which must be solved simultaneously. Since the round-trip time of the gas is in the order of milliseconds, while experimental data is collected over hours, the various stages are assumed to take place instantaneously (that is, no time delay is incorporated to allow for the flowing gas). Because of the time assumption, the three equations could be written in terms of a single differential equation to be solved. The validity of the approach is discussed after presentation of the experimental results (see Discussion and standard physical chemistry text<sup>8</sup>).

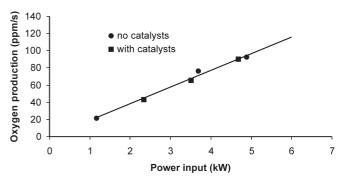


Fig. 7. Trace of oxygen production rate versus power input and model prediction.

## Discharge only

Experimental data suggest that the rate at which  $O_2$  is generated within the discharge is independent of both the gas mixture used and the gas temperature, and depends only on input energy into the discharge. However, some recombination takes places as the hot gas mixes, even when no catalyst is present, as is evident from the fact that the oxygen levels within the laser do stabilize at some point, albeit at an unacceptably high level. In this model only stages 1 and 2 were considered, and the coupled systems written as a single differential equation of the form:

$$\frac{dx(t)}{dt} = k_d - k_g x(t),\tag{1}$$

with solution

$$x(t) = \left(x_0 - \frac{k_d}{k_g}\right) \exp(-k_g (t - t_0)) + \frac{k_d}{k_g}.$$
 (2)

Here x(t) is  $[O_2]$  as a function of time, measured in parts per million (ppm);  $k_d$  is the oxygen generation rate within the discharge, in units of  $\rm s^{-1}$ ; and  $k_g$  is the self-catalytic rate converting the oxygen back to carbon dioxide, in units of  $\rm s^{-1}$ . The equation is written in terms of the starting conditions of the gas mix, where  $x_o$  = amount of oxygen at time  $t_o$ . The value of  $k_g$  is very small, and consequently the second term on the right-hand side of Equation (1) only becomes significant at large oxygen concentrations. Experimentally, one finds that  $k_d = cP_{in}$ , where  $P_{in}$  is the power deposited into the discharge (see Fig. 7), and  $c = 19\,310\,{\rm ppm/J}$  or  $\sim 19\,{\rm ppm/s}$  per kilowatt of input power. The data collected with catalysts present fits the trend-line, as expected if  $k_d$  is not dependent on gas species.

## Catalysts present

With catalysts present in the laser, the oxygen concentration stabilizes quickly and at lower levels than in the case of no catalysts, due to the contribution from stage 3 of the system. Using the model already found for the discharge chemistry [Equation (2)], and allowing for stage 3 to drive preferentially the oxidation of CO back to  $\mathrm{CO}_2$ , we can write the coupled system equations as:

$$\frac{dx(t)}{dt} = k_d - k_g x(t) - k_c x^n(t), \tag{3}$$

where we have kept the form of Equation (1), but added an additional term [last term on the right-hand side of Equation (3)] to allow for faster convergence to lower oxygen levels. This additional term is negative, implying that its presence causes oxygen concentrations to decrease in value with time. By trial and error the rate law that worked the best for the catalysts under consideration was the simple case for n = 1, with the solution:

$$x(t) = \left(x_0 - \frac{k_d}{k_g + k_c}\right) \exp(-(k_g + k_c)(t - t_0)) + \frac{k_d}{k_g + k_c}.$$
 (4)

The catalysts discussed in this paper were supplied by STC Catalysts Inc. They were Pt/SnO<sub>2</sub> based, with 2% Pt (±140 g per catalyst) and of honeycomb structure (dimensions 124.5 mm imes129.5 mm  $\times$  50.0 mm). They have the quoted advantages of low pressure drop (400 cpsi) and room temperature operation, making them suitable for use inside a TEA CO<sub>2</sub> laser. Eight such catalysts were used in a grid one layer deep (1120 g Pt in a total honeycomb weight of 4 kg). The gas composition inside the laser was measured at position 4 (see Fig. 6) with a gas chromatograph for general gas species detection, as well as a Teledyne O<sub>2</sub> detector for oxygen detection. The Teledyne system was used as an on-line monitor, giving real-time data on the change in oxygen concentrations with time. The detector was connected to a data acquisition system in order to synchronize the oxygen measurement with that of all the other laser parameters (for example, pulse energy).

It was assumed implicitly in the development of the model that CO,  $O_2$  and  $CO_2$  all react in stoichiometric ratios. This assumption allowed the coupled differential equations to be written in terms of oxygen only. This assumption was tested and the results indicated that the  $CO:O_2$  ratio was  $2.1 \pm 0.07$ , in good agreement with the stoichiometry value of 2. Furthermore, observations of water vapour, ozone and  $NO_x$  gases (using random testing with the gas chromatograph as well as suitable Drager tubes) indicated minimal concentrations throughout the tests, with only the  $NO_x$  levels measurable at 75 ppm, although without catalysts ozone was measured at 50 ppm.

The model was then tested against an arbitrary initial condition of a running laser, followed by a period when the laser is switched off (no discharge), and then switched on again (discharge on) and allowed to stabilize. The test is comprehensive because it requires the model to describe both discharge and non-discharge conditions, as well as both transient and steady-state periods of operation. The success of the model is shown in Fig. 8, where the model prediction (solid curve) shows excellent agreement with the experimental data (data points). Similar agreement is found between experiment and theory when no catalysts are present [Equation (2)].

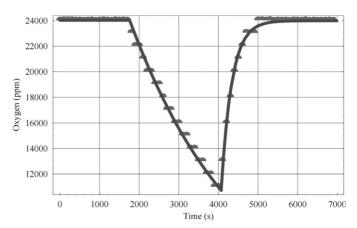
# **Model predictions**

One can use the model to make certain predictions as to the behaviour of the gas in the presence of catalysts. Although it is useful to know the time evolution of the oxygen concentration, as given by Equation (4), one is usually interested in predicting the final steady-state concentrations of oxygen in the system, since this is the level at which the laser will be expected to operate at for extended time periods. If the value is too high, the laser will be unstable in operation (e.g. due to a high arc rate or a low output energy). The model can also be used to investigate what initial conditions are necessary to ensure stable operating regimes.

Allowing  $t\rightarrow\infty$  in Equation (4), the predicted oxygen levels at steady state are given by

$$[O_2]_{eq} = \frac{k_d}{k_g + k_c(T)} \approx \frac{19P_{in}}{k_c(T)}.$$
 (5)

Here  $P_{in}$  must be expressed in kW, and we have written the catalyst rate constant as a function of temperature in anticipation of the analysis to follow (note that replacing  $k_c$  with  $k_c(T)$  does not influence any of the other results in any way). At first inspection it appears that, as expected, increasing the power into the discharge,  $P_{in}$ , increases the final steady-state oxygen level, while improving the efficacy of the catalysts,  $k_c$ , decreases the steady-state level (since the system is dynamic, if the rate at which oxygen is removed is increased, the steady state value also changes).



**Fig. 8.** The model prediction (solid curve) of oxygen levels as a function of time measured during transient and steady-state conditions indicating excellent agreement with the experimental data (solid disks).

For greater insight one needs to understand what influences  $k_c$ ; the major contribution discussed here is temperature. Noting that it is generally assumed that rate constants follow the Arrhenius law, we can write:

$$k_c(T_g) = A \exp\left(-\frac{E_a}{RT_g}\right),\tag{6}$$

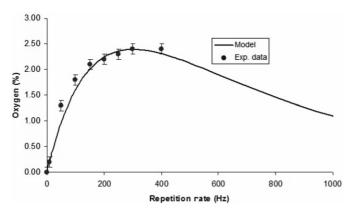
where  $E_a$  is the activation energy of the reaction, A is a constant, R is the universal gas constant and  $T_s$  is the temperature at which the reaction takes place, which is assumed to be equal to the temperature of the gas entering the catalysts. The temperature of the gas at the catalysts can easily be calculated from basic thermodynamics, and can be shown to be given by:

$$T_g = T_0 \left( \frac{P_{in}R}{P_0 I dv C_v} + 1 \right), \tag{7}$$

where *l* and *d* are the electrode length and gap, respectively, *v* is the gas speed through the electrodes, and  $C_v$ ,  $T_0$  and  $P_0$  are the heat capacity, temperature and pressure, respectively, of the gas entering the discharge. In this model some of the gas is heated by passing through the discharge area where a large amount of energy is deposited into the gas. Note that not all the gas experiences the discharge environment since the laser is pulsed at a specified repetition rate and thus not all the gas is heated. This is taken into account in Equation (7). During flow to the catalysts, the heated gas mixes with the unheated gas, and thus the temperature  $T_g$  at the catalysts is lower than the temperature of the gas immediately after the discharge. Now both  $k_d$  and  $k_c$  are functions of the power input to the discharge:  $k_d$  directly, and  $k_c$ through the fact that an increase in power causes an increase in gas temperature, as shown in Equations (6) and (7). The question can be asked: do oxygen levels ever decrease with increasing input power? Combining Equations (5), (6) and (7) indicates that they do, with the turning point in power given by:

$$P_{\text{max}} = \frac{P_0 I dv C_v}{R T_0} \left( \frac{E_a - 2RT_0 - \sqrt{E_a (E_a - 4RT_0)}}{2R} \right). \tag{8}$$

It is interesting to note that the empirically determined constant in the relation  $k_d \approx 19~P_{in}$  does not appear in this equation. A plot of this prediction is shown in Fig. 9, together with experimental data. In this experiment, the final steady-state oxygen levels were measured as a function of the input power to the laser. For practical reasons only, the input power to the laser was achieved through increasing the laser repetition rate (pulses per second), since the input power to the laser is simply the



**Fig. 9.** Steady-state oxygen levels (experiment and theory) as a function of the laser repetition rate (where input power is proportional to the laser repetition rate).

product of the energy deposited per pulse into the discharge and the number of times per second this is done. Unfortunately, the laser under consideration could not reach high enough repetition rates to test the prediction thoroughly. However, there is generally good agreement between the theory and the experiment within the region tested, which is encouraging.

#### Discussion

The model was based on a three-step approximation to a continuous flow system, with recycling gas, which resulted in a dynamic system described by coupled differential equations. The 'guesses' at the rate laws governing the three stages were determined by extensive experimental data on time-dependent concentration changes using the half-life method, as well as non-linear least-squares fits to the data. The validity of the model assumptions is evident from the excellent agreement with experimental data, as well as the confirmation of predictions made with the model. The model correctly predicts both the transient nature of the system under study, as well as the steady-state conditions. What is not understood is why this model works so well. It is a simple approximation to a very complicated catalytic process. Unreported data suggests that the rate law for the catalysts depends on [CO], and it is suspected that water content also plays a significant role in this relationship. The catalyst stage alone could in fact be written as a system of many coupled

Long-term data over more than a billion pulses shows a more complicated relationship between energy and oxygen concentrations<sup>9</sup> than depicted in these experiments. It is thought that over long time frames the presence of excess oxygen has a deleterious effect on the laser, thus influencing the output energy through several mechanisms (e.g. corrosion of the pre-ionization system, which reduces energy). The catalyst activity at the start of the tests was comparable to that of good

catalysts quoted elsewhere, but had degraded to only 30% of that activity by the end of the first 1 billion pulses. This, coupled to the high discharge generation rate of  $O_2$ , resulted in very high steady-state levels of oxygen: 2.4% after 1.3 billion pulses compared to 0.7% at the start of the tests.

Figure 9 suggests a least favourable operating condition for the laser, corresponding to the energy input associated with the peak oxygen concentration. This suggests that for this particular laser to operate at lower oxygen levels, the catalysts would have to be considerably more effective. Monitoring of the catalyst behaviour over extended periods showed a marked decrease in catalyst activity, which resulted in a decreased energy output from the laser.

### Conclusion

We have completed a design and optimization process on a TEA  $\mathrm{CO}_2$  laser to be used in LU. The results show that for LU applications, lower output coupler reflectivities and gas mixes with high  $\mathrm{CO}_2$  content are preferred. The process included a detailed study of the chemistry within the laser, and the impact this has on laser performance over time. The outcome of the study was a final laser parameter configuration suitable for use in LU. The final laser design is presently under manufacture at Scientific Development and Integration, a South African technology company, where contract research and development continues. The prototype system discussed in this paper is in operation at a Lockheed Martin aerospace company for LU testing of modern jet fighters.

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