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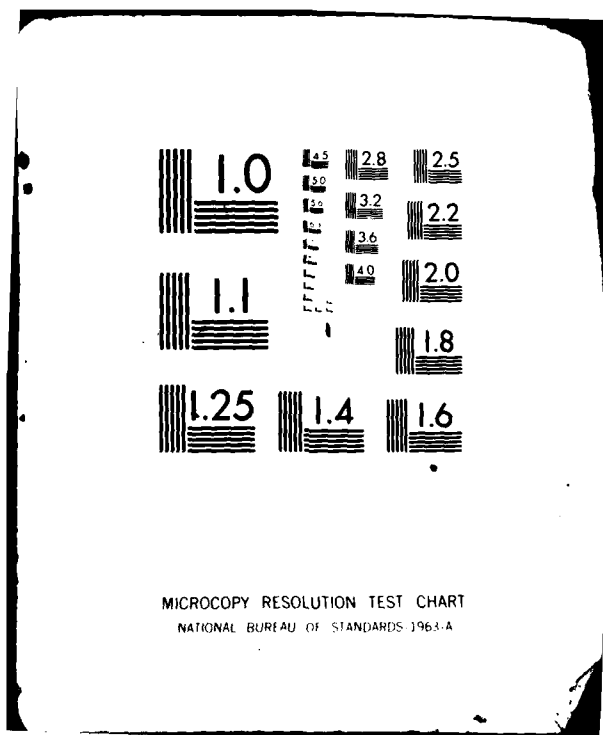
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PARTICLE BEAM CHEMISTRY STUDIES

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Calculations were made of the chemistry in the channel left by a high intensity particle beam pulse. It was found that, because of the high temperature of the gas in the channel, the electron density was determined almost exclusively by electron-ion recombination. The role of nitrogen vibration in determining the temperature was also investigated.		

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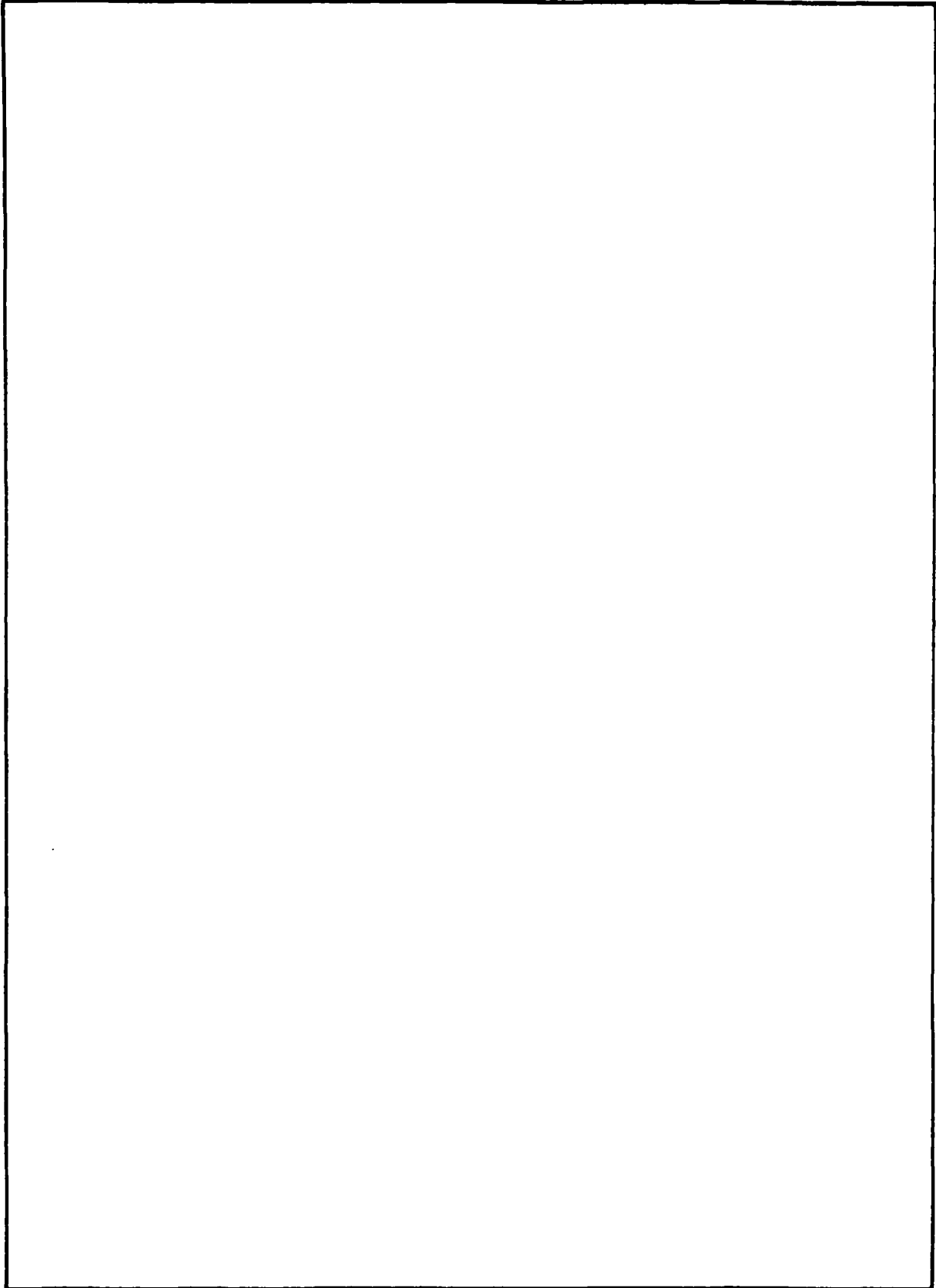
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PARTICLE BEAM CHEMISTRY STUDIES

INTRODUCTION

The successful propagation of a pulsed particle beam depends in part on the conditions in the air channel which is formed. In particular, the conductivity is a critical parameter, and this is determined primarily by the electron density. This report describes a short study which consisted of several chemistry calculations in the channel following an intense 2 mm radius beam and one calculation of the chemistry in the beam.

CHANNEL INITIAL CONDITIONS

The temperatures and species concentrations at the end of the beam pulse were taken from a calculation made by SAI¹ for a 10 kA, 2 mm radius pulse. Although the pulse length was 10^{-8} sec, the SAI calculation was carried out to 2×10^{-8} sec because of considerable ohmic heating which occurs after the high energy particle beam passes. The conditions at 2×10^{-8} sec were used as initial conditions in our calculation. The heavy particle temperature was 1810K, the electron temperature was 6380K and the nitrogen vibrational temperature was 5642K. The species concentrations are shown in Table 1.

The case chosen by SAI was a particularly intense beam and involves the deposition of a considerable amount of energy. The total energy deposited, including ohmic heating, is about 3.5×10^7 ergs/cm³. About thirty percent of this energy is contained in N₂ vibration and thirty percent in dissociation and electronic excitation. This energy can be released by chemical reactions and excited state quenching during the

Table 1. Species concentrations at 2×10^{-8} sec.

E	=	3.06 (15)*
N ₂	=	1.97 (14)
N ₂ (A)	=	7.60 (16)
O ₂	=	4.58 (18)
O ₂ (¹ Δ)	=	2.76 (17)
O ₃	=	2.03 (12)
N	=	4.17 (17)
N(² D)	=	5.17 (16)
O	=	1.02 (18)
O(¹ D)	=	6.01 (14)
NO	=	1.70 (17)
NO ₂	=	2.16 (14)
N ⁺	=	2.36 (12)
N ₂ ⁺	=	8.60 (12)
N ₃ ⁺	=	1.04 (13)
N ₄ ⁺	=	4.82 (11)
NO ⁺	=	2.34 (15)
O ⁺	=	4.63 (10)
O ₂ ⁺	=	8.32 (14)
O ⁻	=	4.45 (13)
O ₂ ⁻	=	8.82 (13)

* Number in parenthesis is the power of ten.

evolution of the channel and can significantly impact the gas and electron temperatures.

CHANNEL EXPANSION

The heating of the gas will cause an expansion of the channel. The initial heating of the gas to 1810K will cause an expansion (assuming adiabatic expansion, vibrational freezing and no further energy release) of about a factor of four and a concurrent cooling of the gas to about 1070K. This expansion takes several microseconds to occur but since we will be interested in tens of microseconds we have assumed the expansion takes place instantaneously. We therefore reduced all the species concentrations in Table 1 by a factor of four and used these values as our starting concentrations. In addition we made an estimate of the N_2O which would have formed during the energy deposition and added this to the initial conditions. This was done because N_2O , at these temperatures, is an excellent catcher of electrons. The value of N_2O we assumed after expansion was $3.6 \times 10^{15} \text{ cm}^{-3}$.

CHEMISTRY CALCULATIONS

The chemical evolution of the channel was calculated using our SEACHEM code. This code integrates the coupled differential chemical equations and contains about 70 species and about 600 reactions. Since the chemical development will involve the release of energy stored in dissociation and vibration during the energy deposition period, the code keeps track of the energetics and allows for heating of the gas. It also allows for instantaneous adiabatic expansion of the gas as heating occurs. This density decrease amounted to factors of between 1.5 and 2 in our calculations, depending on the assumptions.

We will not list all the reactions in our code. As it turned out the high temperatures in the channel simplify the chemistry and only several reactions or processes are critical to the electron density.

When the ionization source turns off at 10^{-8} seconds the dominant ion is O_2^+ . Because of charge exchange reactions NO^+ quickly becomes the dominant ion thereafter and by 10^{-7} seconds is the only significant positive ion. The electron-ion recombination rate of NO^+ is therefore an important rate constant. We have used a value of

$$k_1 = 4 \times 10^{-7} \left(\frac{300}{T_e} \right) \text{ cm}^3/\text{sec} \quad , \quad (1)$$

where T_e is the electron temperature. There is, however, considerable uncertainty in the dependence on T_e . Experimental values range from $T_e^{-.4}$ to T_e^{-1} . For an electron temperature of about 0.5 eV or 6000K, this amounts to a difference in the rate constant of a factor of six. In a recombination dominated decaying plasma this amounts to a factor of six difference in the electron density.

Electron attachment is usually an important parameter in determining the electron density. The main attachment mechanism is usually three body attachment to O_2 , i.e.,



The production of N_2O , however, in large amounts makes the following reaction potentially important,



Although this reaction is endothermic by about 0.2 eV, the presence of both hot electrons and vibrationally excited N_2O can make the rate constant for reaction 3 significant. Using the experimental data² and an electron temperature of several tenths of an eV we have obtained the rate constant

$$k_3 = 10^{-8} \left(\frac{T}{300} \right) e^{-3200/T} \text{ cm}^3/\text{sec}, \quad (4)$$

where T is the N₂O temperature.

Detachment will also be important and the primary detachment mechanisms are



Reaction 4 is important because of the high temperatures involved in the channel.

Ion-ion recombination is usually dominated by the three-body process at sea level density and temperature. Because of the heating and expansion of the channel, however, the two-body process must be included. The effective two body rate constant used in our calculations was

$$k_9 = 6 \times 10^{-8} \left(\frac{300}{T} \right)^{0.5} + 10^{-25} \left(\frac{300}{T} \right)^{2.5} (M) \text{ cm}^3/\text{sec} \quad (9)$$

where T is the gas temperature and (M) is the total particle density in cm⁻³.

Finally, because of the energy stored in N_2 vibration, the quenching of N_2 vibration is an important parameter. Atomic oxygen seems to be the most efficient quencher of N_2 vibration of all the species involved in our calculations. The measured rate constants between 1000K and 2000K can be fit by the expression³

$$k_{10} = 7.8 \times 10^{-15} \left(\frac{T}{300} \right)^{1.7} \text{ cm}^3/\text{sec} \quad (10)$$

A question which arises, however, is how efficient is the quenching by oxygen of the high vibrational states of N_2 . If the quenching of the high vibrational states can also be given by the above expression then the N_2 will lose a significant portion of its energy to translation in several tens of microseconds. If not it will take significantly longer.

RESULTS

Four calculations were made. In two of the calculations the N_2 vibration was assumed to be quenched slowly while in the other two the quenching was assumed to be rapid. Since the interchange of energy between electrons and nitrogen vibration is very rapid⁴ we assumed that the electron temperature was equal to the N_2 vibrational temperature. Thus the quenching of N_2 controls the electron temperature. In each pair of calculations one calculation assumed no water vapor and the other assumed a 3 percent H_2O concentration. The H_2O was added to see if this made a significant difference in the electron density due to clustering of the positive ions and the formation of ions such as H_2O^+ and H_3O^+ . In addition, the inclusion of H_2O will yield species such as H , OH and HO_2 . These species can greatly effect the oxygen chemistry and therefore impact on the nitrogen vibrational quenching.

The electron densities for these four cases as a function of time (in microseconds) are given in Table 2. Cases A and B are for minimum N_2 vibrational quenching. Case A is for no H_2O and case B is for 3 percent H_2O . Cases C and D are for maximum N_2 vibrational quenching, with case C having no H_2O and case D, 3 percent H_2O .

Table 2. Electron densities.

t(μ sec)	Case A	Case B	Case C	Case D
10	2.1 (12)*	2.1 (12)	1.8 (12)	1.9 (12)
20	1.1 (12)	1.1 (12)	1.1 (12)	1.1 (12)
40	5.9 (11)	6.1 (12)	6.4 (11)	6.3 (11)
70	3.8 (11)	3.8 (11)	3.5 (11)	3.1 (11)
100	2.9 (11)	2.7 (11)	2.1 (11)	1.9 (11)

* Number in parenthesis is power of ten.

We see that there is very little variation in the electron density among the different cases except at late times and then only by, at most, 50 percent. The addition of water vapor makes no significant difference in the type of ion which is present. Because of the high temperatures involved there is no clustering and NO^+ remains the dominant ion. The addition of H_2O does make a significant difference in the recombination of atomic oxygen. The atomic oxygen disappears much more rapidly and this does decrease the N_2 quenching. However, part of this decrease is offset by quenching by H and H_2 . Also the more rapid recombination of oxygen adds to the heating of the gas and this offsets the reduced gas heating caused by the reduced N_2 vibrational quenching. A more accurate

treatment of the interaction between the electron temperature and N_2 vibration would probably make the differences between the water and no water cases larger but they would still not be significant.

The presence of N_2O does increase the attachment significantly at early times. At late times the high temperature has caused much of the N_2O to thermally dissociate. Also at late times the attachment, which is still large even without the N_2O , is offset by an equally large detachment caused in large part by the high temperature. Thus the electron density decay is electron-ion recombination dominated. The electron density is essentially determined by the dissociative recombination rate constant for NO^+ .

CALCULATION WITHIN THE BEAM

The addition of water vapor in the previous calculations had no effect in changing the fact that NO^+ was the dominant ion. However, the SAI calculation within the beam had no water vapor. The ions N_2^+ , O^+ and N^+ can charge exchange with water to form H_2O^+ and this ion can react with water to form H_3O^+ . We thought that the H_3O^+ , which has a larger recombination rate than NO^+ , formed within the beam might persist and contribute to the electron decay after the beam had passed. We therefore made a calculation within the beam. The ion-production rate specified by SAI was used (about 4×10^{17} ion-pairs/cm³/sec for 10^{-8} seconds) as was their results for the gas and electron temperatures. The calculation showed that H_2O^+ and H_3O^+ were indeed significant within the beam but that these species decayed rapidly after the ionization source is turned off. By 2×10^{-8} seconds, the recombination rate was totally dominated by NO^+ and O_2^+ .

This calculation, however, did show significant differences in the concentrations of a number of other species at 2×10^{-8} seconds between

the SAI calculation and ours. Our atomic oxygen concentration is about half that obtained by SAI and this is due to H and OH chemistry. Our $O_2(^1\Delta)$ concentration was only 40 percent the corresponding SAI value and this was probably due to our inclusion of quenching by electrons and perhaps a lower value of the $O_2(^1\Delta)$ formed per initial ion-pair. These reduced values could impact on later detachment and N_2 vibrational quenching, particularly in less intense, lower temperature beam cases.

Our $N(^2D)$ concentration was about an order of magnitude smaller than the SAI value and this is undoubtedly due to the quenching of this state by H_2O . This quenching probably causes the species NH to be formed and this causes a whole new chemistry problem to be introduced since the chemistry of NH is largely unknown.

Our value of ozone was considerably larger than that in the SAI calculation. We obtained an O_3 concentration of $8.7 \times 10^{14} \text{ cm}^{-3}$, which was about 400 times the SAI value. This large formation was due to the detachment reactions



This may be somewhat unrealistic since the reverse of these reactions were not included even though they are only about 0.6 eV endothermic. The rate constants, however, cannot be obtained from detailed balance since these reactions almost certainly involve O_3 in vibrationally excited states. The dependence of the rate constants on the vibrational states is unknown and must be determined experimentally.

Finally the electron density we obtained at 2×10^{-8} seconds was $8.3 \times 10^{14} \text{ cm}^{-3}$, a factor of about four less than that in the SAI

calculation. This is due to the attachment to N_2O formed in our calculation. As we have already pointed out, however, this N_2O will be destroyed later by the high temperature.

CONCLUSIONS

The main conclusion to be drawn from our calculations is that the chemistry of very intense beams is fairly simple due to the high temperatures involved. The electron concentration is primarily determined by the electron-ion recombination rate constant for NO^+ . Unfortunately this rate constant is highly uncertain at high temperatures. We used a T_e^{-1} dependence. Use of a $T_e^{-0.4}$ dependence would probably reduce the electron density to below 10^{11} cm³/sec after 40 microseconds. This could have serious consequences for the stability of the second pulse trying to propagate in the channel at those times.⁵ Another uncertainty resides in the electron temperature at microsecond times due to the uncertainty involved in the quenching of N_2 vibration and its coupling to T_e . This should be examined more carefully.

There is considerable interest in less intense beams.⁵ In these the heating will be much less severe and the chemistry in the channel will be more complicated. Cluster ions will be important and will persist out to late times. Attachment will be greater due to N_2O and probably O_3 . This should be investigated in more detail. Detachment will be much less. The quenching of N_2 vibration will be reduced and this will keep the electron temperature higher for a longer period of time. Previous calculations have indicated that it will take about three pulses to produce a highly ionized channel ($n_e > 10^{11}$). However, in these calculations ohmic heating was not included, T_e was wrong, N_2 vibration was not included, the expansion was not properly handled and the chemistry scheme was not as complete.

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