

Chemistry and Physics of Weakly Ionized Plasmas

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Final Report

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14. ABSTRACT The goal of this task is to make measurements for plasma chemistry over a wide range of temperatures, pressures and other conditions. We have succeeded in pushing the boundaries for conditions in a number of ways. Our ranges of conditions include the highest temperature measurements for both ion molecule and electron molecule reactions. We have made the first measurements of the products of ion – ion recombination, we have found the first four-body electron-ion recombination reaction and discovered a process we call electron catalyzed mutual neutralization. We have also developed techniques to handle excited oxygen, and to study electron attachment to radicals. We have also used what we have learned from the above studies for trace gas detection using chemical ionization mass spectrometry. The summary of the work is below and those interested can look at the list of 131 publications for more specific details					
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1. INTRODUCTION

During the course of this task we have studied numerous aspects of plasma chemistry. The main results can be found in the 131 publications listed below. There are four main areas of plasma chemistry and we have made significant contributions in all of them during this task. Originally our expertise involved ion-molecule chemistry and to a lesser extend electron molecule chemistry. During the course of this task, we have also made substantial progress in understanding electron-ion recombination and ion-ion mutual neutralization. We have applied the results to chemical ionization mass spectrometric measurements of trace gases in jet and rocket exhaust, volcanos and the atmosphere. The results have been transition to a variety of Air Force users involving the ionosphere, reentry, plasma assisted combustion, and electric laser plasmas. This report will highlight this progress by plasma reaction type in bullets.

2. ION MOLECULE REACTIONS

Reactions of $S_mF_nO^-$ with O_3 have been examined. Spin effects are found to be important. $S_mF_nO^-$ thermochemistry was explored by a combination of laboratory and ab initio calculations. Problems with the electron affinity ladder were found. The results are applicable to SF_6 atmospheric releases and transformers which use SF_6 as a discharge suppressant.

Mobilities of mass selected cluster ions were studied for the first time showing problems with previous measurements made elsewhere without mass selection.

The branching ratios for the reaction of O_2^+ with C_8H_{10} show a strong dependence on pressure in agreement with our previous hypothesis derived from low pressure chemistry.

A study of the reaction of HCl^+ with CF_4 has resolved a long standing debate on the thermochemistry of CF_3^+ . The new measurements show the reaction proceeds by proton transfer followed by thermal decomposition instead of dissociative charge transfer as previously thought.

Two papers have been published on the ion and electron chemistry of the atmospherically important molecule, SF_5CF_3 . They show that charged particles may contribute to the atmospheric lifetime of this molecule.

A number of ionospheric reactions have been measured at high temperature. This continues previous work on ionospheric chemistry at high temperature. Results of these and previous ionospheric kinetics are being parameterized and incorporated into models of the natural and artificial (HAARP) ionosphere. The results indicate that the separation of energy into translational, rotational, and vibrational energy effects is important.

Studies of the stabilization of the charge transfer products of alkylbenzenes reacting with O_2^+ have been completed. Data has been combined with statistical analysis from Jurgen Troe to

derive the energy transferred per collision. These are the first measurements of this quantity for ions.

Measurements of N_x^+ ($x=1-3$) with NO reactions have been made over wide temperature ranges. Except for a small channel involving N_2^+ production in the N^+ reaction, all reactions proceed through charge transfer. In all cases rotational and translational energy have been found to be equivalent, consistent with previous studies on other systems. Vibrational energy has either no effect or a negative effect on the reactivity.

We have studied the reactions of $PO_xCl_y^-$ with O_2 , O_3 , H_2 , and H. Previous flame studies showed that introducing $POCl_3$ into a flame produces mainly PO_2^- and PO_3^- . The present results show, for the first time, a pathway from the products of electron attachment to $POCl_3$ to PO_x^- .

The first accurate kinetics for reactions of negative ions at high temperature have been made. Reactions of CO_3^- and O_3^- with SO_2 have been studied up to 1440 K. This was facilitated by adding a corona discharge ion source to the instrument.

We studied the reaction of N^+ with O_2 with respect to product states. The results showed that little excited NO^+ is formed but that $N(^2D)$ is the primary neutral formed in charge transfer. That has important implications for ionospheric NO production.

Reactions of O and N atoms have been made. The first study of O_2^- reactions showed problems with previous studies. This is important to the electron balance in the D-region.

Chemistry involving $PO_xCl_y^-$ and $SO_xF_y^-$ ions reacting with O and N has been studied. Pathways to PO_2^- and PO_3^- have been found that help explain Gooding's flame chemistry.

Much effort has gone into studying reactions involving $O_2(a^1\Delta_g)$. Our first system studied involved generating $O_2(a)$ by microwave discharge on O_2 . This created O and O_3 impurities and corrections to the data were needed. Using this technique, O_2^- and O^- rate constants were studied at 298 K and it was shown that previous measurements were in error. Subsequently, a sparger chemical generator was used to form $O_2(a)$ with good success. Quantitative conversion of a Cl_2 flow into a flow of O_2 (sum of $O_2(a)$ and $O_2(X)$) occurred in the bubbler. Rate constants for the reaction of $O_2(a)$ with O_2^- and O^- from 200-700 K and with $PO_xCl_y^-$ and $SO_xF_y^-$ ions at 298 K have been measured. We studied numerous negative ion reactions with $O_2(a^1\Delta_g)$. A lot of work went into measuring the ionospheric positive ions reacting with $O_2(a^1\Delta_g)$. They were problematic because they reacted with ground electronic state O_2 and also reacted rapidly with H_2O produced by the sparger. Interestingly, some of the reactions showed that $O_2(a^1\Delta_g)$ reacted more rapidly than does the ground state, and other reactions showed the opposite. In collaboration with Bierbaum and Troe, energy transfer dissociation of negative ion clusters was observed.

We found isomers of $\text{NO}^+(\text{H}_2\text{O})_{3,4}$ for the first time for this important ionospheric system. The collaboration included theoretical groups of McCoy, Jordan, and Tully, and the results have been published in *Science*.

3. ELECTRON ATTACHMENT

Rate constants were measured for electron attachment to $\text{c-C}_4\text{F}_8$, $\text{C}_5\text{F}_5\text{N}$, $\text{C}_5\text{HF}_4\text{N}$, $\text{C}_6\text{F}_5\text{Cl}$, $\text{C}_6\text{F}_5\text{Br}$, $\text{C}_6\text{F}_5\text{I}$, $\text{SF}_5\text{C}_6\text{H}_5$, $\text{SF}_5\text{C}_2\text{H}_3$, and *o*-, *m*-, and *p*-trifluoromethylbenzonitrile, over the temperature range 298-550 K. Several of these have electron affinities low enough that rate constants for thermal electron detachment could be measured. Analysis of the equilibrium allowed the electron affinities to be determined. A new plasma decay effect was observed in the $\text{C}_6\text{F}_5\text{Cl}$ case.

The electron attachment to POCl_3 has been studied in detail. Pressure dependences have been measured and modeled over wide ranges in collaboration with Jürgen Troe. The FALP was upgraded to allow measurements as a function of electron temperature and the first study involved both branching ratios and kinetics for attachment to POCl_3 .

New data from our laboratory and modeling by Dr. Troe for electron attachment to SF_6 has shown that much of the previous data on this extremely important system has been taken under ill defined conditions and therefore is not adequate for modeling correctly. The new data and modeling now show a consistent picture of the reactivity including new measurements of the electron affinity and bond strength.

The high temperature flowing afterglow was upgraded to include a Langmuir probe for studying electron reactions. Numerous problems were solved and electron attachment to NF_3 , CF_2Cl_2 , CF_3Cl , CH_3Cl , and Cl_2 was studied to 1100 K, the highest temperature at which an attachment reaction was studied. The first four molecules showed typical Arrhenius behavior over the whole temperature range. The latter showed bi-exponential Arrhenius behavior. Theoretical analysis by Ilya Fabrikant showed that the behavior was predictable by considering individual vibrational states of Cl_2 and excellent agreement between theory and experiment was found. In addition, attachment to SO_2Cl_2 , SOCl_2 , SOF_2 , SO_2F_2 , and SO_2FCl have been measured to 900 K. Several of the reactions show an unusual dihalide product and thermal dissociation occurs for several ion products.

The turbulent ion flow tube (TIFT) has been used to set an extremely low lower limit for the rate constant for the reaction of O_2^+ with N_2 of around $10^{-21} \text{ cm}^3 \text{ s}^{-1}$. This reaction would be important in the chemistry of the ionosphere at the previous known limit. It is probably the lowest value measured for any ion-molecule reaction rate constant.

4. ELECTRON RECOMBINATION

Electron recombination rate measurements were made at the University of Stockholm. They show a large fraction of recombination occurs by producing 3 neutrals. These will be incorporated into Air Force combustion models. The complete $C_2H_n^+$ series has been studied. Higher values of n produce more of the channel with 2H loss. The data shows the average number of radicals produced is about 2, double that previously incorporated into Air Force combustion models.

The electron recombination of $Na^+(D_2O)$ has been studied as a function electron energy. This ion is important in loss of Na^+ ions in the atmosphere and in reentry. The measurements show that the ion readily recombines and that the clustering of H_2O to Na^+ is the rate controlling step in loss of Na^+ in the atmosphere and reentry.

The analysis of $C_2H_5^+$ recombination shows that a channel with four products ($C_2H_2 + 3 H$) occurs in 13% of the reactions. This is the first time four products have been observed in dissociative recombination except for weakly bonded clusters.

Data on dissociative recombination (DR) for hydrocarbon ions have been combined and correlations made with thermodynamics. This allows predictions of both branching ratios and rate constants for unknown systems. More data are needed to confirm the trend.

Electron recombination of CF_n^+ ions has been studied as a function of electron energy. These measurements are needed to understand reentry involving Teflon heatshields and etching plasmas.

We are currently developing analysis software to look at complicated kinetics in the FALP and HT-FALP. This should allow for studies of attachment to radicals produced in a primary attachment and information on ion-ion mutual neutralization products. We have preliminary data on attachment to CCl_3 , $PSCl_2$ and $PSCl$.

5. ION-ION MUTUAL NEUTRALIZATION

A new technique for studying ion-ion recombination has been developed. It requires adding an electron attaching gas(es) to the FALP where 2 or more ions are formed. By measuring the branching ratios at the end of the flow tube as a function of initial electron density, accurate relative recombination rates with Ar^+ can be derived. If one of the negative ions produced is monatomic, absolute rates are also obtained. A *Mathematica* model of the complete chemistry yields information on both the rates for recombination and the neutral products as well as rates for e^- attachment to the neutral radicals. We have concentrated on recombination of SF_6^- , which requires knowledge of SF_5^- and SF_4^- recombination. This was the first complete product distribution measured for a mutual neutralization reaction of this complexity. More dissociation was observed with SF_6^- due to its lower detachment energy. Theoretical calculations by Jürgen

Troe indicated that the process occurred in two steps: neutralization at long range, followed by energy transfer at the repulsive wall.

6. CHEMICAL IONIZATION MASS SPECTROMETRY

The measurements of gas-phase HNO_3 , SO_2 and HCN in the lower arctic stratosphere made during the winter of 1999-2000 as part of NASA's SOLVE campaign have been analyzed. The HCN measurements are amongst the first in-situ measurements of HCN in the atmosphere and can be used to calibrate remote sensing measurements. The HNO_3 measurements show that renitrification occurs in late winter in the lower stratosphere, in agreement with other measurements that show denitrification at higher altitudes. This has important implications for the Arctic ozone hole.

Measurements of SO_x exhaust gases were made in FY02 behind a B-757 on a runway in conjunction with the NASA atmospheric effects of aviation program.

Our data have been used to show a number of important effects including (1) HNO_3 deposition from polar stratospheric cloud "rain", (2) that ion induced nucleation is important in the upper troposphere/lower stratosphere, and (3) determining SO_2 lifetimes after an volcanic eruption.

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