**Reactions of \(N^+\), \(N_2^+\), and \(N_3^+\) with NO from 300 to 1400 K**

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**Abstract**
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Reactions of $N^+$, $N_2^+$, and $N_3^+$ with NO from 300 to 1400 K

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Rate constants have been measured from 300 to 1400 K in a selected ion flow tube (SIFT) and a high temperature flowing afterglow for the reactions of $N^+$, $N_2^+$ and $N_3^+$ with NO. In all of the systems, the rate constants are substantially less than the collision rate constant. Comparing the high temperature results to kinetics studies as a function of translational energy show that all types of energy (translational, rotational, and vibrational) affect the reactivity approximately equally for all three ions. Branching ratios have also been measured at 300 and 500 K in a SIFT for the $N^+$ and $N_2^+$ reactions. An increase in the $N_2^+$ product at the expense of NO nondissociative charge transfer product occurs at 500 K with $N^+$. The branching ratios for the reaction of $N_3^+$ with NO have also been measured in the SIFT, showing that only nondissociative charge transfer giving NO+ occurs up to 500 K. The current results are discussed in the context of the many previous studies of these ions in the literature. [DOI: 10.1063/1.1792232]

I. INTRODUCTION

Measurements of the kinetics of ion-molecule reactions at high temperatures have proven that the internal energy available at these elevated temperatures can have a significant effect on the rate constants and product branching ratios. These data show that extrapolations to high temperature behavior based solely on room temperature data could be seriously in error. The results have been invaluable in modeling the chemistry at elevated temperatures involving plasmas.

The reactivities of the nitrogen ion species $N^+_m$ where $m = 1–3$ are of particular interest. Both $N^+$ and $N_2^+$ are key precursors in the complex chemistry of the ionosphere and $N_3^+$ ions are readily formed in rf discharges. In addition, NO+ ions are one of the main terminal ions in air plasma environments and are a major component of the daytime ionosphere of altitude of 200 km, making reactions with NO especially interesting.

Considering the importance of the ion-molecule chemistry of nitrogenic cations with NO, the rate constants for the reactions of $N^+$, $N_2^+$, and $N_3^+$ with NO have been studied up to 1400 K using a selected ion flow tube (SIFT) and a high temperature flowing afterglow (HTFA). Previous drift tube studies of the kinetics as a function of reactant translational energy have been done at 298 K for all three ions. Thus, comparing the current results with the drift tube data illustrates the effects of internal energy on the reactivity. In addition, the temperature dependence of the branching ratios for the reactions of $N^+$ and $N_2^+$ have been measured up to 500 K in the SIFT where the reactant ions are mass selected. The products for the $N^+$ reaction have previously been measured only at 298 K, indicating that another reactive channel occurs in addition to charge transfer. However, the product branching ratios for the $N_3^+$ reaction have not been measured previously. For each individual ion-molecule reaction, numerous previous studies using several different methods will be discussed to better understand the current results.

II. EXPERIMENT

Rate constants for the $N^+$, $N_2^+$, and $N_3^+$ reactions with NO have been measured using both a SIFT and HTFA at the Air Force Research Laboratory. The instruments have been described in detail elsewhere, including later modifications to the upstream ion source region of the HTFA. The helium buffer gas was passed through a liquid nitrogen cooled sieve trap to remove water vapor. The NO was also passed through a sieve trap to reduce the minute amount of impurities in the sample. In the following sections, only the details pertinent to the current experiments will be discussed.

In the SIFT, electron impact on nitrogen gas taken from the boiloff of a high pressure liquid nitrogen dewar and introduced into an effusive source produced $N^+$ ions in a high pressure source chamber. $N_3^+$ ions were simultaneously generated by the three-body reaction of $N^+$ with $N_2$ and Penning ionization of $N_2$ with $N_2^+$ in the source. The ion of interest was mass selected with a quadrupole mass analyzer and injected into a fast flow of helium buffer gas (AGA, 99.997%) that was introduced through a Venturi inlet. These ions became thermally equilibrated before entering the reaction zone where NO gas (AGA, 99.8%) was added.

After reaction over a known distance at a previously measured reaction time under pseudo-first-order conditions, the remaining ions were sampled through an aperture in a blunt nose cone, mass analyzed, and then detected. The product distribution was determined by extrapolating the branching ratios to zero NO flow to minimize the effects of secondary reactions with NO in the flow tube, leading to relative...
errors in the branching ratios of ±10% of the major product peaks.\textsuperscript{19} The experimental rate constants have relative uncertainties of ±15% and absolute uncertainties of ±25%.\textsuperscript{19}

In the HTFA, the three nitrogen cations of interest were generated simultaneously without mass selection in an ion source region upstream of and perpendicular to the main flow tube. The helium buffer flow was introduced into the source region through an inlet behind a thoriated filament. Electron impact on the buffer gas generated He\textsuperscript{+} and He\textsuperscript{2+} ions. These ions reacted with nitrogen gas introduced downstream from the filament to generate N\textsuperscript{+} and N\textsuperscript{2+} ions. The N\textsuperscript{3+} ions were generated through the same two mechanisms as in the SIFT. The pressure in the source region was increased by the use of a 0.25 in. diameter diaphragm so that the N\textsubscript{2} concentration in the flow tube was low enough to minimize the clustering reaction there. The various ions were transported into the flow tube by the fast flow of buffer gas where a commercially available furnace heated the flow tube to the desired temperature. These ions became thermally equilibrated before entering the reaction zone where the NO was introduced. Again, after reaction over a known distance at a previously measured reaction time under pseudo-first-order conditions, the remaining ions were sampled through an aperture in a blunt nose cone, mass analyzed, and then detected. The experimental rate constants measured with the HTFA have relative uncertainties of ±15% and absolute uncertainties of ±25%.\textsuperscript{20}

III. RESULTS

A. N\textsuperscript{+} + NO

The reaction of N\textsuperscript{+} with NO has been extensively studied in the past.\textsuperscript{11,12,16–18,23–27} Rate constants for this reaction are shown in Fig. 1 plotted against temperature. For the drift tube data measured in a flow drift tube (FDT) and selected ion flow drift tube (SIFDT),\textsuperscript{12} the center-of-mass kinetic energies have been converted to an effective translational temperature $T_{\text{eff}}$, where $T_{\text{eff}}=2\left(KE_{\text{cm}}\right)/k_B T$ and $k_B$ is Boltzmann’s constant. The rate constants decrease with increasing temperature and can be represented by a power law as $(6.5\times10^{-9})T^{-0.44}$. The room temperature rate constants shown in Fig. 1 measured in various fast flow tube instruments\textsuperscript{12,17,18,26} as well as an ion cyclotron resonance (ICR) mass spectrometer\textsuperscript{16} are in good agreement, having a value of ca. $5.5\pm0.7\times10^{-10}\text{cm}^3\text{s}^{-1}$. Two measurements of the rate constant are not shown in Fig. 1 because they were made in the early days of the flowing afterglow, before error levels had been reduced. Those measurements gave somewhat larger values of $8\sim9\times10^{-10}\text{cm}^3\text{s}^{-1}$.\textsuperscript{23,24} The average value of the room temperature rate constant is approximately one-half of the Su-Chesnavich collision rate constant\textsuperscript{28,29} of $1.0\times10^{-9}\text{cm}^3\text{s}^{-1}$. The new SIFT and HTFA data are in agreement with the drift tube data\textsuperscript{12} up to 900 K within the 15% relative error. However, the new data above 1000 K are lower than the corresponding drift tube results\textsuperscript{12} for the reasons described below.

The SIFT and HTFA rate constants reflect thermal energy distribution of the reactants at the flow tube temperature, which covers the range from 300 to 1400 K for the current measurements. A survey of numerous past high temperature studies of the kinetics of ion-molecule reactions shows that having energy in either reactant translation or rotation usually affects the reactivity equally.\textsuperscript{1,30} To investigate the role of rotational and translational energy, particularly at high temperatures, the rate constants are alternatively plotted versus the sum of the average rotational and translational energy in Fig. 2. The average rotational energy of the NO is simply $k_B T$, where $T$ is the reaction temperature, and the average kinetic energy in the SIFT and HTFA is simply...
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\[ N⁺ + NO \rightarrow NO⁺(X) + N + 5.3 \text{ eV}, \]
\[ \rightarrow N₂⁺(X) + O + 2.2 \text{ eV}, \]
\[ \rightarrow O⁺ + N₂ + 4.2 \text{ eV}. \]

The predominant product observed in the SIFT is nondissociative charge transfer yielding NO⁺, accounting for 0.91 of the products at 300 K and 0.88 of the products at 500 K. The previous values are in the 0.79–0.93 range and the 300 K data are in closest agreement with the ICR value of Anicht, Huntress, and Futrell. The other major reaction channel gives products N₂⁺ and O, where the N₂⁺ branching fraction increases from 0.07 at 300 K to 0.11 at 500 K. This is outside our error of 20% of the minor channel. Again, the new 300 K value agrees best with the ICR result. Creating either the N₂⁺(A) or O(1D) excited neutral products is 1.1 and 0.21 eV exothermic, respectively. The relatively small amount of N₂⁺ may indicate barriers along this reactive pathway or competition with charge transfer. Fahey et al. have also examined the possibility of forming O⁺ and N₂ products at high kinetic energies and place an upper limit on the O⁺ branching ratio of 0.05 from 0.05 to 1.5 eV. The present values show a minor O⁺ product with a branching fraction of 0.02 and 0.01 at 300 and 500 K, respectively, consistent with the earlier observations. The O⁺ product can also be exothermically produced in the 2D electronic excited state.

Tichy et al. have found the highest fraction of N₂⁺ products (0.21) in a SIFT experiment that probed the reactivity of both ground and excited state N⁺ using monitor ion techniques.17 The reaction of N*⁺ with H₂ gives H₂⁺ provides a measure of the reactivity attributable to the metastable ions present. However, the authors note that N⁺ reaction with H₂ complicates the deconvolution of the quenching of N*⁺ relative to reaction. N⁺ reacts rapidly with H₂ to give NH⁺ which, in turn, reacts rapidly with NO to give N₂H⁺ at m/z = 29.18 If the product mass analyzer resolution is insufficient to resolve 1 amu, then the N₂⁺ contribution may be artificially inflated in the Tichy et al. SIFT results. Adams et al. have measured a branching ratio of 0.15 for N₂⁺ in a SIFT twice the value obtained currently. Adams et al. have noted that the source conditions have been carefully established to minimize the production of metastable N⁺ ions so the presence of excited species should not interfere with their measurement. No obvious explanation can be found for the discrepancies between the current and previous results. All of the N_m⁺ ions are present in the HTFA during these measurements, precluding the measurement of branching ratios.

B. N₂⁺ + NO

The reaction of N₂⁺ with NO has also been extensively studied by various methods.10,13,14,17,23–26,34–43 The rate constants for this reaction are plotted as a function of temperature in Fig. 4. Again, the data follow a power law dependence, \( k = (7.5 \times 10^{-9})T^{-0.52} \). The dependence of the rate constant for N₂⁺(v = 0) on the center-of-mass kinetic energy (KE_c.m.) has been measured in two separate SIFDT experiments using a monitor gas to isolate the N₂⁺ ground vibrational state kinetics. For clarity, the Innsbruck SIFT results of Dobler et al.13 are labeled SIFTD and the SIFDT results of Howorka, Albrighton, and Fehsenfeld14 measured at NOAA are labeled NOAA in Fig. 4 and subsequent figures. Cross sections for this reaction as a function of (KE_c.m.) measured in a static DT by Kobayashi and Koneko10 and in a guided-ion beam (GIB) by Graul et al.42 have been converted to rate constants by taking the product of the cross section and the average relative velocity of the reactants in the center of mass. All of the (KE_c.m.) from the previous experiments10,13,14,42 have been converted to an effective temperature as described above for N⁺. An additional crossed beam experiment by Turner, Rutherford, and Stebings has probed the cross sections as a function of kinetic energy for this reaction.25 However, their measurements have been made at kinetic energies of 4 eV and higher, which exceeds the overlapping range of the experiments shown in Fig. 4 and have consequently not been shown.

The rate constant at 298 K for N₂⁺ reacting with NO is well established at around \( 4.0 \pm 1.0 \times 10^{-10} \text{ cm}^3 \text{s}^{-1} \).10,13,14,23,24,26,34–38,43 Again, this average value is roughly half of the Su-Chesnavich collision rate constant28,29 of \( 8.5 \times 10^{-10} \text{ cm}^3 \text{s}^{-1} \). The current HTFA results agree with the previous studies showing that the rate constants decrease with increasing temperature. The two SIFDT studies13,14 agree well the HTFA data up to 900 K. The HTFA rate constant measured at 1000 K appears out of line with the other values. Varying the HTFA reaction conditions including filament emission current, N₂ source gas flow, sampling voltage, detector voltages, lens conditions, and electron energy did not affect the rate constant at 1000 K, eliminating the most commonly found sources of...
error. In spite of these tests, we believe the shape of the data curve indicates that there must be a small systematic error in that point. The DT data agree with the flow tube results at 298 K and both the GIB and DT data exhibit the same qualitative trend. However, the DT and GIB data are systematically higher. We have no obvious explanation for the discrepancy. However, our past experience has proven that data obtained with the HTFA and obtained with the NOAA and Innsbruck drift tubes are generally in excellent agreement. Therefore, in order to derive internal energy dependences, we compare the HTFA results only to the SIFDT and NOAA data.

To investigate the role of translational and rotational energy at increased temperatures, the rate constants are also plotted against the average translational and rotational energy in Fig. 5. Only the HTFA, SIFDT and NOAA data are plotted for clarity. The average rotational energy for NO in all of the experiments is \( k_B T \), where \( T \) is the temperature of the experiment. The average rotational energy in the \( N_2^- \) in the drift tube experiments is \( k_B T_{\text{buf}} \), where \( T_{\text{buf}} \) is determined from the center-of-mass collision energy for ion-buffer gas collisions \( \langle KE_{\text{buf}} \rangle = 3/2 k_B T_{\text{buf}} \).

The HTFA, SIFDT (Ref. 13), and NOAA (Ref. 14) data sets in Fig. 5 agree within the error in the experiments, intimating that translational and rotational energy behave similarly. The HTFA data are lower than the drift tube data at temperatures over 1000 K but just at the limit of our uncertainty estimate. This discrepancy could be due to vibrational excitation reducing the reaction rate. However, it would mean that the \( v > 0 \) rate constants would be effectively zero and therefore, the difference is more likely a result of a small systematic error in one of the measurements. In any case, it is clear that vibrational excitation does not significantly enhance the rate constants. For \( N_2^- \) vibrations, it is shown that the rate constant for reactivity is similar between various states and that the overall rate constant increases because of quenching.

The reaction of \( N_2^- \) with NO proceeds almost exclusively via charge exchange to give \( NO^+ \). Three energetically possible channels are illustrated below in Eq. (2).

\[
\begin{align*}
N_2^- + NO &\rightarrow N_2 + NO^+ (X) + 6.3 \text{ eV}, \quad (2a) \\
&\rightarrow N_2 + NO^+ (a) - 0.07 \text{ eV}, \quad (2b) \\
&\rightarrow N_2 (A) + NO^+ (X) + 0.15 \text{ eV}. \quad (2c)
\end{align*}
\]

At collision energies above 5 eV in the GIB, two dissociative charge transfer channels giving \( N^+ \) and \( O^+ \) have been observed when these pathways become energetically accessible as shown in Eq. (3).

\[
\begin{align*}
N_2^- + NO &\rightarrow N_2 + N^+ (S) + O^+ (S) - 4.6 \text{ eV}, \quad (3a) \\
N_2^- + NO &\rightarrow N_2 + N^+ (3P) + O^+ (3P) - 5.5 \text{ eV}. \quad (3b)
\end{align*}
\]

Production of the \( NO^+(a) \) in the HTFA experiments could cause the apparent rate constants for reaction (2) to be slightly smaller since the \( NO^+(a) \) ions would charge transfer back to \( N_2^+ \). However, no curvature in the HTFA data indicative of this back reaction has been observed, nor do the rate constants depend on the \( N_2 \) flow rate. As discussed previously, product branching ratios could not be obtained in the HTFA because all three of the \( N_2^+ \) ions are present under the current experimental conditions.

As previously mentioned, around 10% of the \( N_2^+ \) vibrational population is in \( v = 1 \) at 1400 K. The quenching of vibrationally excited \( N_2^+ \) by NO in competition with the charge transfer reaction has been studied previously.\(^{13,14,17,38,40,41}\) Most recently, Frost et al.\(^{43}\) have used a SIFT with laser-induced fluorescence detection of the \( N_2^+ \) vibrational levels to probe the competing processes. As found previously,\(^{41}\) the \( v = 1 \) levels decay at a faster rate than \( v = 0 \). However, ca. 70% of the reaction from \( v = 1 \) is charge transfer giving \( NO^+ \) with a rate constant only 10% larger than that for \( v = 0 \).\(^{43}\) The HTFA results show that all types of energy affect the energy essentially the same, in keeping with the results of Frost et al.\(^{43}\)

### C. \( N_2^+ + NO \)

Unlike \( N^+ \) and \( N_2^+ \), only one previous study of the rate constant for the reaction of \( N_2^+ \) with NO has been performed.\(^{15}\) Lindinger has studied the kinetic energy dependence of the rate constant in a FDT at two different flow tube pressures of 298 K using a nitrogen buffer. The rate constants as a function of temperature are plotted in Fig. 6, where the \( \langle KE_{\text{e.m.}} \rangle \) from the FDT is converted to \( T_{\text{tr}} \) as discussed above. All of the data sets shown in Fig. 7 agree well, except at 500 K where the SIFT value is slightly lower than the FDT results. However, the rate constants agree within the combined error of the two experiments. The rate constants are much less than the Su-Chesnavich collision rate constant of \( 7.7 \times 10^{-19} \text{ cm}^3 \text{ s}^{-1} \). Nevertheless, given the excellent agreement as a function of temperature between the FDT and HTFA, the data can be compared as a function of total en-
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\[ N^+ + NO \rightarrow N + NO \]

**Energy** as shown in Fig. 7 to examine the role of the total energy available. The translational, rotational, and vibrational energy in \( N_3^+ \) and NO are calculated similarly as for the \( N_2^+ \) reaction. An important difference is that the vibrational temperature of the \( N_3^+ \) in the FDT reflects \( T_{\text{buf}} \). When plotted this way, the HTFA data in Fig. 7 are lower than the FDT data at energies above 0.4 eV; however, the rate constants still agree within the error of the two experiments. One major difference is that the FDT experiments have been done in an N\(_2\) buffer.\(^{15}\) Therefore, the ion velocity distribution will be different from similar measurements in a helium buffer and the remaining differences may be attributable to the difference in the energy distribution using \( N_2 \) as the buffer gas in the drift tube.\(^{46}\)

Lindinger\(^{15}\) proposes that \( N_2O^+ \) products may be formed exothermically in the \( N_3^+ \) reaction.\(^{31}\) However, only nondissociative charge transfer giving NO\(^+\) has been observed in the SIFT at 300 and 500 K as shown in Table I and illustrated in Eq. (4) below. The heat of formation of \( N_3^+ \) used to calculate the reaction enthalpy is an average of data taken from the literature,\(^{31,47-49}\)

\[ N_3^+ + NO \rightarrow NO^+ (X) + N_2 + N(4^1S) + 1.7 \text{ eV} \]  

(4)

Forming linear \( N_3 \) products intact instead of dissociated as shown above is also exothermic,\(^{31,47-49}\) however \( N_3 \) is unstable. The branching ratios have not been measured previously\(^{15}\) and are unable to be examined in the HTFA for reasons outlined above.

**IV. DISCUSSION**

Figure 8 shows all of the energy dependent kinetics data measured using flow tubes for \( N_m^+ + NO \) with \( m = 1-3 \) plotted together versus the average total energy available. All of the drift tube data are plotted in the top graph and all of the HTFA data are in the bottom graph. The overall rate constant for a given ion decreases with an increasing number of N atoms in the cation. However, the \( N_3^+ + NO \) rate constants show a marked increase with increasing energy above ca. 0.25 eV in both the drift tube and the HTFA. The Franck-Condon factors at the charge transfer energy resonance with NO based on the recombination energies\(^{51}\) of the reactant ions for \( N^+ \) (14.53 eV), \( N_2^+ \) (15.58 eV), and \( N_3^+ \) (11.27 eV) producing the NO\(^+\)(\(X\)) state are not favorable for any of the
ions. The mechanisms below are complicated and several mechanisms may play a role which may account for variations in the reactivity as seen in Fig. 8.

The trends in the rate constants with energy are indicative of reactions involving a collision complex. N+ reacting and higher in the GIB. The internal energy appears to be more varied than in the reactivity as seen in Fig. 8. The mechanisms below are complicated and several have been found through measuring product kinetic energy distributions using an ICR that the rate constant for reaction 1 is a function of the internal energy. The internal energy appears to be the same for reactions involving the same neutral reactant.

A more direct mechanism may contribute to the N+ + NO reaction as well. The strongest Franck-Condon overlap would be at the resonance with the NO+(X)+N(2P) products, where the product curve crosses at around NO(v = 4), but this crossing would form a large barrier to charge exchange along this coordinate. It is possible that the entrance channel potential could shift relative to the product potential as the reactants approach each other. This region of the potential would be controlled by the global minimum of the NO surface instead, possibly creating resonances with the N+ ground state, making this pathway less likely over the energy range presently studied. In addition, the potential surfaces explicitly for the N+ + NO system are unknown. Thus, the topology of the surface for the N+ reaction may be different for the N+ reaction.

V. CONCLUSIONS

This work continues our series of studies on how different forms of energy affect the rate constants for ion-molecule reactions. Previously, we have shown that in almost all cases, rotational and translational energy affect the reactivity equally within our error. All three reactions studied here confirm that observation. The effects of vibrational energy have been more varied. We have previously concluded that for charge transfer reactions, the effects of vibrations are often the same for reactions involving the same neutral reactant. This was certainly true for O2 reactions, where O2 vibrational excitation enhanced the reactivity. However, for NO reactions, only two have been studied up to 1400 K and no appreciable effect of vibrationally excited NO was observed. All three reactions shown here are consistent with this observation. However, the large vibrational constant for NO results in only a small amount of excited NO, making accurate determination of the vibrational rate constants impossible. In any case, those rate constants cannot be appreciably larger than the ground state rate constant and are probably very similar. This conclusion is consistent with the Franck-Condon factors being large if the lowest NO+(X)
vibrational levels are populated. All of the reactions with NO studied here involve a collision complex, consistent with the fact that redistribution of energy results in all forms of energy behaving the same. Curve crossings in the potential energy surface at long range for the systems involving NO may also have an influence on the observed reactivity.

The temperature dependence of the branching ratios for the N⁺ reaction with NO has also been measured for the first time up to 500 K. An increase in the amount of N₂ products at the expense of NO⁺ nondissociative charge transfer products occurs at 500 K. A small amount of O⁺ product is also observed. The branching ratios for the reaction of N₂ with NO have also been measured in the SIFT, showing that only nondissociative charge transfer to give NO⁺ occurs up to 500 K.

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