Electronic emission due to collisions involving low energy
CHO$^+$ and H$^+$ ions and CH$_4$ and N$_2$ molecules

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Abstract. Electronic emission from CH and OH radicals, CH$^+$, CO$^+$ and N$_2^+$ ions and
atomic hydrogen lines have been observed during collisions of CHO$^+$ and H$^+$ ions with
CH$_4$ and N$_2$ molecules in the laboratory kinetic energy range 100-900 eV. Emission cross
sections of the excited CH radicals, N$_2^+$ ions and H$_\alpha$ transition of H atoms have been
measured in the above energy range.

1. Introduction

Ion–molecule reactions involving collisions of ground and/or excited state species
have been a subject of increasing interest in the past (Bowers 1986, Fontijn 1985,
Franklin 1972), mainly because of their direct involvement in the complex
chemistry/physics of fuel-rich hydrocarbon flames, interstellar chemistry, discharge
and laser-initiated plasmas, etc. The chemi-ionization processes have been predicted
to be the leading cause of soot formation in oxy–hydrocarbon flames. Ions such as
CHO$^+$ and C$_3$H$_5^+$ have been postulated to be the root cause of soot formation in
fuel-rich flames. According to Calcote (1981), the CHO$^+$ ions are formed due to
associative ionization of CH(X)+O→CHO$^+$+e with a rate constant of $k =
2.3 \times 10^{-13}$ cm$^3$ mol$^{-1}$ s$^{-1}$ in the temperature range 2000-2400 K (Peeters and Vinckier
1975). Here CH(X) is the ground-state of the CH radical. However, Phippen and
Bayes (1989) and Bayes (1988) have recently shown that involvement of the metastable
CH(a$^2 \Sigma$) rather than the ground state CH(X) radical is a strong possibility in the
formation of CHO$^+$ ions. The CH(a$^2 \Sigma$) states lies $\approx 0.69$ eV above the ground state
CH(X $^2 \Pi$) (Neils et al 1988). Cool and Tijssen (1984) have shown enhancement of
the above rate constant by a factor of over 2000 with electronically excited state
CH(A $^2 \Delta$) or CH(B $^2 \Sigma$) radicals. In a hydrocarbon–air flame, a large number of ground
state as well as excited state CH radicals (in addition to many other radicals such as
C$_2$, OH, etc) are expected to be observed and have, in fact, been observed by many
previous investigators (Calcote 1981, Peeters and Vinckier 1975, Miller 1976, Le and
Vanpee 1985). Therefore, a large number of CHO$^+$ ions are expected to be present in
the flame front. In addition to CHO$^+$ ions, other ions such as H$_2$O$^+$ and C$_2$H$_5^+$ have
also been observed in flame fronts and their concentration is many times greater than
the concentration of CHO$^+$ ions (Calcote 1981). The loss of the CHO$^+$ ions is a direct
result of fast ion–molecule reactions in the flame medium. At a flame temperature of
2000-4000 K, several processes such as chemi-excitation, chemi-ionization, ion–
molecule reactions, ionization due to collisions of excited state species and thermal
electrons, superelastic collisions involving hot electrons and excited state atoms, molecules and ions, may be responsible for the formation of the ionized species. It is then clear that flames may not be the best experimental medium to study the role played by the many individual chemical and physical processes that make up the total flame. For a better understanding of the flame chemistry, it is desirable to study these processes individually, preferably at room temperature.

The CHO⁺ ions have also been observed in interstellar molecular clouds (Buhl and Snyder 1970, Herbst et al. 1976, Duley and Williams 1984). There are two isomers of the CHO⁺ ion with potential energy minima on the (H-C-O)⁺ surface (Woods et al. 1975, Gudeman and Woods 1982). In the interstellar clouds, these isomers are believed to be produced due to the reactions: H₃⁺ + CO → HCO⁺ (or HOC⁺) + H₂ and CO⁺ + H₂ → HCO⁺ (or HOC⁺) + H. The HCO⁺ is produced in greater concentration and is known to be more stable than HOC⁺. Indeed, the number density of HCO⁺ ions overwhelms HO⁻ ions by the ratio 94:6 in a given reaction involving the two isomers (Illies et al. 1983, Redeker et al. 1985). The ion–molecule reactions involving collisions of CHO⁺ ions and CH₄ molecules have been studied at thermal energies. Adams et al. (1978) observed no reaction byproducts and placed a limiting value on the total rate constant \( k \leq 1 \times 10^{-12} \text{ cm}^3 \text{ mol}^{-1} \text{ s}^{-1} \), whereas Roche et al. (1971) have observed CH₃⁺ + CO as the byproducts with a rate constant \( k \leq 1 \times 10^{-11} \text{ cm}^3 \text{ mol}^{-1} \text{ s}^{-1} \). In a recent study, Adams et al. (1991) have studied the dissociative recombination CHO⁺+e and have quantitatively determined the OH product distribution using laser fluorescence techniques. The total dissociative recombination rate coefficients for the CHO⁺+e reaction have been measured by Leu et al. (1973) at room temperature as well as at 205 K and are equal to \((3.3 \pm 0.5) \times 10^{-7} \text{ cm}^3 \text{ mol}^{-1} \text{ s}^{-1}\) and \((2.0 \pm 0.3) \times 10^{-7} \text{ cm}^3 \text{ mol}^{-1} \text{ s}^{-1}\), respectively. The rate coefficient measured by Amano (1990) for the same process at 273 K is \(3.1 \times 10^{-7} \text{ cm}^3 \text{ mol}^{-1} \text{ s}^{-1}\).

Based on the above description, it is clear that there is no information available on the electronically or vibrationally excited states of the byproduct species formed during collisions of CHO⁺ ions and electrons or neutral species. In this communication, we wish to report our results on the electronic excitation of several species observed during collisions of CHO⁺ and H⁺ ions with O₂, N₂, H₂ and CH₄ molecules.

2. Experimental procedure

The experimental set-up used to collect data reported here has been described in detail elsewhere (Kushawaha and Michael 1988, 1989) and will not be repeated here. However, a brief description of the procedure used to collect the data will be given here. The projectile ions CHO⁺ and H⁺ were generated by electron impact with flowing methanol vapour and extracted through a small hole in the anode. The ions were then accelerated and passed through a magnetic mass selector and subsequently through a collision cell containing the desired target species. After passing through a collision cell, these ions were collected on a Faraday cup and measured by a sensitive electrometer. The target gas pressure was measured by a calibrated pressure gauge.

The light emitted from the collision cell due to the ion–molecule reaction inside the collision cell was viewed in a direction perpendicular to the ion beam. The light photons were dispersed by a 0.2 m scanning monochromator and detected by a cooled (-25 °C) photomultiplier tube (PMT). The signal from the PMT was amplified and sent to a multichannel analyser for storage, analysis and subsequent plotting of the data.
Electronic emission spectra from atomic hydrogen, i.e. H\(_0\), H\(_\alpha\), and H\(_\beta\) lines, CH and OH radicals and CH\(^+\), N\(_2^+\) and CO\(^+\) ions, were observed and recorded in the wavelength range 580-240 nm during collisions involving CHO\(^+\) and H\(^+\) as projectile ions and CH\(_4\), H\(_2\), and N\(_2\) molecules as target species. The intensities of the atomic and molecular features were observed to be different at different laboratory kinetic energies of the projectile ions. The emission spectra were recorded only in the laboratory kinetic energy range of 100-900 eV. At kinetic energies lower than 100 eV, the ion current was too small and the spectra were too weak for calculating emission cross sections and a meaningful interpretation of the data was not possible. No attempt was made to record the emission spectra above the laboratory kinetic energy of 900 eV, mainly because of experimental limitations. During the collisions of CHO\(^+\) ions with CH\(_4\) molecules, strong emission from the electronically excited states of CH radicals, i.e. CH (A \(^2\Delta\) - X \(^2\Pi\)) and CH(B \(^2\Sigma^-\) - X \(^2\Pi\)), OH radicals due to the transition (A \(^2\Sigma^-\) - X \(^2\Pi\)), and CH\(^+\) and CO\(^+\) ions due to transitions CH\(^+\) (A \(^1\Pi\) - X \(^1\Sigma^+\)) and CO\(^+\) (B \(^2\Sigma^+\) - X \(^2\Sigma^+\)), respectively, and atomic lines of hydrogen were observed. The intensity of the Q-head of the (0, 0) band due to the transition CH(A \(^2\Delta\) - X \(^2\Pi\)) was observed to be the strongest compared with the other band head due to this transition or other emission features observed due to CHO\(^+\) and CH\(_2\) reactions under given experimental conditions. When CH\(_4\) molecules were replaced by N\(_2\) molecules, strong emission was observed from the (0, 0), (1, 0) and (2, 0) band heads due to the transition N\(_2^+\) (B \(^2\Sigma^+\) - X \(^2\Sigma^+\)) as compared with the CH(B \(^2\Delta\) - X \(^2\Pi\)) and OH (A \(^2\Sigma^-\) - X \(^2\Pi\)) radicals. Atomic lines of hydrogen were completely absent in this reaction. The emission due to the CH\(^+\) ions was either not present in the spectrum or it was completely overlapped by the strong (0, 1) band head of the N\(_2^+\) ion due to the transition (B \(^2\Sigma^+\) - X \(^2\Sigma^+\)). The same trend continued in the kinetic energy range of 100-900 eV at a particular pressure of the N\(_2\) gas. With either CH\(_4\) or N\(_2\) molecules and the CHO\(^+\) ion, no emission spectrum due to CO molecules was observed at higher kinetic energies of the projectile ions, i.e. higher than 500 eV. However, at kinetic energies below 500 eV, a very weak emission spectrum, probably due to CO molecules, was observed in the wavelength range 570-480 nm. No electronic emission, atomic or molecular, was observed when H\(_2\) or O\(_2\) gas was substituted for CH\(_4\) or N\(_2\) gas and the experiments were performed with CHO\(^+\) ions in the laboratory kinetic energy range 100-900 eV. When CHO\(^+\) ions were replaced by H\(^+\) ions and reactions were studied with CH\(_4\) molecules as target gas, strong emission lines of hydrogen atoms and emission bands due to CH radicals and CH\(^+\) ions were observed. The emission bands due to the OH radical transition (A \(^2\Sigma^-\) - X \(^2\Pi\)) was also observed as an impurity. In figure 1, we display the emission spectra observed due to collisions of CHO\(^+\) and H\(^+\) ions with CH\(_4\) and N\(_2\) molecules at a laboratory kinetic energy of 900 eV. The emission bands denoted by I in figure 1 (upper scan) are probably due to N\(_2^+\) as an impurity. These peaks may be compared to the (1, 0) and (2, 0) bands due to the N\(_2^+\) band heads observed during the collisions of CHO\(^+\) ions and N\(_2\) molecules shown in the middle scan of figure 1. The band head
Figure 1. Electronic emission spectra observed due to collisions of various ions and molecules at a laboratory kinetic energy of 900 eV and CH₄/N₂ pressure of 4 mTorr. The spectra are not corrected for the optical detection efficiency. Upper scan, CHO⁺ + CH₄; middle scan, CHO⁺ + N₂; bottom scan, H⁺ + CH₄. H₂, H₂, and H₂ are the atomic hydrogen lines at 486 nm, 434 nm and 410 nm, respectively. The (0, 0), (1, 0) and (2, 0) are the band heads due to N₂⁺(B 2Σ⁺-X 2Σ⁺) transition. A, P-head of the (0, 1) band of the CH(A 3Δ-X 3Π) transition; B, Q-head of the (2, 2) band of the CH(A 3Δ-X 3Π) transition; C, Q-head of the (0, 0) band of the CH(A 3Δ-X 3Π) transition; D, Q-head of the (0, 0) band of the CH(A 3Δ-X 3Π) transition; E, R-head of the (0, 0) band of the CH⁺ (A 1Π-X 1Σ⁺) transition; G, CO⁺ band due to the transition (B 2Σ⁺-X 2Σ⁺) transition; OH, R₂-head of the OH-radical due to the (A 2Σ⁺-X 3Π) transition.

due to the CH (B 2Σ⁺-X 3Π) transition and denoted by D in the upper scan of this figure may be slightly overlapped by the (0, 0) band head of N₂⁺ as shown in the middle scan. The spectrum due to the collision H⁺ + CH₄ is shown as the lowest scan in figure 1 and is free from any N₂⁺ band structure. The assignments of these band heads are from Pearse and Gaydon (1976).

There are two processes which may be responsible for the observation of excited state atomic and molecular emission described above. These processes include collision-induced dissociative charge transfer and excitation processes. For the CHO⁺ + CH₄ collisions, these processes may be written as follows:

\[ \text{CHO}^+ + \text{CH}_4 \]

\[
\rightarrow \text{CHO}(X) + \text{CH}_4^+ + \Delta H = -4.6 \text{ eV} \quad (1)
\]

\[
\rightarrow \text{CHO}^+(B^2\text{A}^+) + \text{CH}_4^+ + \Delta H = -9.56 \text{ eV} \quad (2)
\]

\[
\rightarrow \text{CHO}^+ + \text{CH}(A^2\Sigma) + H_2 + \Delta H = -6.33 \text{ eV} \quad (3)
\]

\[
\rightarrow \text{CHO}^+ + \text{CH}(B^2\Sigma) + H_2 + \Delta H = -6.66 \text{ eV} \quad (4)
\]

\[
\rightarrow \text{CH}(A^2\Delta) + O + \text{CH}_4^+ + \Delta H = -9.01 \text{ eV} \quad (5)
\]
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\[ \rightarrow \text{CH}(B^2\Sigma^-) + \text{O} + \text{CH}_4^+ + \Delta H = -9.34 \text{ eV} \]  \( (6) \)

\[ \rightarrow \text{CO}(X) + \text{H}(n = 3) + \text{CH}_4^+ + \Delta H = -8.41 \text{ eV} \]  \( (7) \)

\[ \rightarrow \text{H}^+ + \text{CO}(B^2\Sigma^+) + \text{CH}_4 + \Delta H = -2.66 \text{ eV} \]  \( (8) \)

\[ \rightarrow \text{H} + \text{CO}^+(B^2\Sigma^+) + \text{CH}_4 + \Delta H = -0.24 \text{ eV} \]  \( (9) \)

\[ \rightarrow \text{OH}^+(A^2\Sigma^+) + \text{CH}(X) + \text{CH}_4^+ + \Delta H = -5.68 \text{ eV} \]  \( (10) \)

where \((X)\) or \((\ast)\) are the ground or excited state species and \(\Delta H\) is the exothermicity or endothermicity of the reaction. Here \(\Delta H > 0\) means the reaction is exothermic. The \(\Delta H\) values for the above processes were calculated by using the ionization \((I)\), dissociation \((D)\) and electronic excitation \((E)\) energies of the various species involved in reactions \((1)-(10)\). The ionization potentials \((I)\) are \(I(\text{CH}) = 10.6 \text{ eV}, I(\text{CH}_4) = 12.70 \text{ eV}, I(\text{CHO}) = 8.1 \text{ eV}, I(\text{CO}) = 14.01 \text{ eV},\) and \(I(\text{CH}_3) = 9.8 \text{ eV},\) the dissociation energies are \(D(\text{CH}) = 3.46 \text{ eV}, D(\text{CH}^+) = 4.08 \text{ eV}, D(\text{CO}) = 11.09 \text{ eV}, D(H\text{-CO}) = 1.54 - 1.0 \text{ eV}\) and the excitation energies are \(E(\text{CH}, A^2\Delta) = 2.87 \text{ eV}, E(\text{CH}, B^2\Sigma^-) = 3.2 \text{ eV}, E(\text{OH}, A^2\Sigma^+) = 4.01 \text{ eV}, E(\text{CO}, B^2\Sigma^+) = 13.2 \text{ eV}, E(\text{CO}^+, B^2\Sigma^+) = 4.95 \text{ eV}\) and \(E(\text{CHO}^+, B^2\Pi) = 4.96 \text{ eV}\). The excitation and dissociation energy values were taken from Huber and Herzberg (1979) and the ionization potential energies were taken from Rosenstock et al (1977), except \(I(\text{CHO})\), which was taken from Dyke (1987). Based on the \(\Delta H\) values calculated using the value of \(I(\text{CHO}) = 8.1 \text{ eV},\) as noted above, most of the processes are highly endothermic, except reaction \((9)\) which is slightly endothermic. Observation of the various highly excited state species clearly indicates that for the endothermic processes to occur some other mechanism(s), completely different from the exothermic processes, is involved for the electronic excitation of these species. The most likely mechanism may be the conversion of the translational energy of the ions into internal energy of the byproduct species. Such a conversion of energy has been observed by us and others in the past for similar ion-molecule reactions for which the endothermicity was more than 6 eV (Kushawaha and Michael 1988, 1989, Kelley et al 1978). A set of similar reaction sequences may be written to explain the observation of the electronically excited state species involving collisions of \(^1\text{H}_2^+\) ions and \(\text{CH}_4\) or \(\text{N}_2\) molecules. At this point, it may be worth noting that the reactions involving \(\text{CHO}^+\) and \(\text{H}_2\) or \(\text{O}_2\) are highly endothermic and produce no emission. It may be possible that conversion of the translation energy into internal energy of the product species did occur but only a small number of excited state species were produced, leading to very weak emitted radiation from the excited state which could not be detected by our detection system.

The emission cross sections of the electronically excited state species was calculated by using the following expression (Kushawaha and Michael 1988, 1989):

\[ \sigma = I_s / I_p n L \]

Here \(\sigma\) is the emission cross section in cm\(^2\), \(I_s\) is the photon counts per second corrected for the detection efficiency of the optical system that included the solid angle of light collection, \(I_p\) the number of ions per second participating in the collision process inside the collision cell, \(n\) the number density of the target species in cm\(^3\) and \(L\) (in cm) is the interaction length of the ion beam and the target gases inside the collision cell. By using the appropriate information in the above expression, we have calculated the emission cross section of the most intense atomic lines or molecular bands observed due to various emitters described above. The results of these calculations are reported in tables 1-3 for \(\text{H}_2\), \(\text{CH} (A^2\Delta - X^2\Pi)\) and \(\text{CH} (B^2\Sigma^- - X^2\Pi)\) emitters in the laboratory.
### Table 1. Emission cross sections of atomic hydrogen line Hβ, CH radicals in the excited states CH(A) = CH(A 2Δ-X 2Π) and CH(B) = CH(B 2Σ-X 2Π) observed during the collisions of CHO+ ions and CH₄ molecules (4 mTorr) in the laboratory kinetic energy range 100-900 eV.

<table>
<thead>
<tr>
<th>Kinetic energy (eV)</th>
<th>Cross section (10⁻²⁰ cm²)</th>
<th>Hβ</th>
<th>CH(A)</th>
</tr>
</thead>
<tbody>
<tr>
<td>900</td>
<td></td>
<td>0.2</td>
<td>1.02</td>
</tr>
<tr>
<td>750</td>
<td></td>
<td>0.8</td>
<td>3.70</td>
</tr>
<tr>
<td>650</td>
<td></td>
<td>1.24</td>
<td>6.32</td>
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<tr>
<td>550</td>
<td></td>
<td>0.76</td>
<td>5.62</td>
</tr>
<tr>
<td>450</td>
<td></td>
<td>0.12</td>
<td>8.70</td>
</tr>
<tr>
<td>300</td>
<td></td>
<td>—</td>
<td>4.40</td>
</tr>
<tr>
<td>100</td>
<td></td>
<td>—</td>
<td>2.74</td>
</tr>
</tbody>
</table>

### Table 2. Emission cross sections of atomic hydrogen line Hβ, CH radicals in the excited states: CH(A) = CH(A 2Δ-X 2Π) and CH(B) = CH(B 2Σ-X 2Π), observed during the collisions of H⁺ ions and CH₄ molecules (4 mTorr) in the laboratory kinetic energy range 100-900 eV.

<table>
<thead>
<tr>
<th>Kinetic energy (eV)</th>
<th>Cross section (10⁻²⁰ cm²)</th>
<th>Hβ</th>
<th>CH(A)</th>
<th>CH(B)</th>
</tr>
</thead>
<tbody>
<tr>
<td>900</td>
<td></td>
<td>2.0</td>
<td>1.6</td>
<td>0.35</td>
</tr>
<tr>
<td>750</td>
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<td>4.4</td>
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<td>8.3</td>
<td>1.87</td>
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<td></td>
<td>7.5</td>
<td>8.7</td>
<td>0.70</td>
</tr>
<tr>
<td>100</td>
<td></td>
<td>4.2</td>
<td>2.0</td>
<td>0.40</td>
</tr>
</tbody>
</table>

### Table 3. Emission cross sections of CH radicals CH(A) = CH(A 2Δ-X 2Π) and (0, 0), (1, 0) and (2, 0) band heads of N₂⁺ (B 2Σ-X 2Σ⁺) transition observed during the collisions of CHO⁺ ions and N₂ molecules (4 mTorr) in the laboratory kinetic energy range 100-900 eV.

<table>
<thead>
<tr>
<th>Kinetic energy (eV)</th>
<th>Cross section (10⁻²⁰ cm²)</th>
<th>CH(A)</th>
<th>(0, 0)</th>
<th>(1, 0)</th>
<th>(2, 0)</th>
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<tr>
<td>900</td>
<td></td>
<td>0.25</td>
<td>1.30</td>
<td>1.69</td>
<td>0.50</td>
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<tr>
<td>700</td>
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<tr>
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<tr>
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<td>—</td>
<td>0.67</td>
<td>1.00</td>
<td>—</td>
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</tbody>
</table>
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kinetic range 100-900 eV. It is to be pointed out here that the cross section for the formative of CH(B) radicals due to collisions of CHO⁻ + CH₄ cannot be measured reliably due to possible overlap by the (0, 0) band head of the N₂⁺ (B-X) system (see figure 1) which may be present as an impurity. These cross sections may be regarded as accurate to within an experimental uncertainty of ±30%. This estimate is based on the measured emission cross sections of atomic hydrogen lines observed due to collisions of He⁺ and H₂ in our laboratory and those measured by Isler and Nathan (1972). The two cross sections were in agreement to within ±20%. This agreement gave us confidence in the procedure used to calibrate our detection system and in the detection efficiency that was used to calculate the values of Iₑ in the above expression. Other details of our calibration procedure are published elsewhere (Kushawaha and Mahmood 1987).

In conclusion, we have studied the ion-molecule reactions involving collisions of CHO⁻ and H⁺ ions with CH₄, N₂, O₂ and H₂ molecules in the laboratory kinetic energy range 100-900 eV. During the collisions, we have observed electronic emission due to CH and OH radicals, CH⁺, CO⁺ and H₂⁺ ions and atomic hydrogen lines. By using the integrated intensity of the strongest bands and atomic lines, we have calculated the emission cross sections in the laboratory kinetic energy range 100-900 eV. These cross sections may be useful in theoretical modelling of the interstellar chemical processes and a better understanding of the chemical/physical processes at work in an oxy-hydrocarbon flame.

Acknowledgments

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