A KINETIC MODEL FOR EXCIMER UV AND VUV RADIATION IN DIELECTRIC BARRIER DISCHARGES*

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AGENDA

• Introduction

• Plasma Chemistry for Xe/Cl₂

• Description of the Model for Microdischarges

• Results for Microdischarge Mechanics and UV Radiation

• Concluding Remarks
Dielectric barrier discharges (DBDs) have shown promise as high intensity sources of UV and VUV radiation emitted by dimer and trimer excimers.

These UV photons have a number of applications:
- Biological sterilization
- Photochemical degradation of organic compounds in flue gases
- Photo-induced surface modification and material deposition
- Lithography

DBDs are usually operated at high pressure on the order of 1 bar, so a number of microdischarge or breakdown channels are generated.
- 2 mm - 5 mm gap
- 10s Hz - a few kHz
EXCIMER GENERATION IN Xe/Cl₂ DISCHARGE

\[ \text{Xe}^* + \text{Cl}_2 \rightarrow \text{Xe}^+ + \text{Cl}^- \]

\[ \text{Xe}^{**} + \text{Cl}_2 \rightarrow \text{Xe}^* + \text{Cl}_2 \]

\[ \text{Xe}_2^* + \text{Cl}_2 \rightarrow \text{Cl}_2^* + \text{Xe} \]

\[ \text{hν} 308 \text{ nm} \]

\[ \text{Xe}^* + \text{e}, \text{Xe}, \text{Cl}, \text{Cl}_2 \rightarrow \text{Xe} + \text{Cl} \]

\[ \text{hν} 172 \text{ nm} \]

\[ \text{Xe}^* + \text{Xe} + \text{M} \rightarrow \text{Xe}_2^* \rightarrow \text{Cl}_2 \rightarrow \text{Xe} + \text{Xe} \rightarrow \text{XeCl}^* \]

\[ \text{Cl}_2^* + \text{e} \rightarrow \text{Cl}^+ + \text{Cl}^- \rightarrow \text{Xe} + \text{Xe} \rightarrow \text{XeCl}^* \]

\[ \text{hν} 259 \text{ nm} \]

\[ \text{XeCl}^* + \text{Xe} + \text{Xe} \rightarrow \text{Xe}_2\text{Cl}^* \rightarrow \text{Xe} + \text{Xe} + \text{Cl} \rightarrow \text{Xe} + \text{Xe} + \text{Cl}_2 \]

\[ \text{hν} 490 \text{ nm} \]
1-d plasma chemistry and hydrodynamics models have been developed to investigate excimer radiation from dielectric barrier discharges in mixtures of xenon and chlorine.
MICRODISCHARGE IN $\text{Xe/Cl}_2 = 99/1$

- $\text{Cl}_2$ has a large rate of dissociative electron attachment at low $E/N$. The electron density shows an expanding avalanching shell for 1% $\text{Cl}_2$.
- An absolute low voltage region is produced in the core of the microdischarge during the voltage pulse, and around the edge after the pulse.

**During Voltage Pulse**

**After Voltage Pulse**

- $\text{Xe/Cl}_2 = 95/1$,
  - Pressure: 0.6 atmosphere,
  - Gas Temperature: 300 K,
  - $V_0 = 10 \text{ kV},$
  - $\tau = 40 \text{ ns},$
  - $\varepsilon = 5 \varepsilon_0$
The formation of the XeCl* excimer is delayed compared to electron generation.

The total generation 308 nm photons is ultimately uniform across the microdischarge.

- Xe/Cl₂ = 99/1, 0.6 atm, 300 K, 
  \( V_0 = 10 \text{ kV} \), \( \tau = 40 \text{ ns} \), \( \varepsilon = 5 \)
OTHER PHOTONS IN Xe/Cl₂ = 99/1

- Xe 2⁺ (172 nm) has a similar lifetime as XeCl⁺.
- Xe 2Cl⁺ (490 nm) has a long lifetime since its quenching rate is independent of the Xe pressure and only dependent on the Cl₂ density.
- Cl₂⁺ (259 nm) can be depleted by conversion to XeCl⁺.

- 1 atm, 400 K, \( V_0 = 12 \text{ kV} \), \( \tau = 40 \text{ ns} \), \( \varepsilon = 5 \)
MICRODISCHARGE IN Xe/Cl₂ = 95/5

- Xe/Cl₂ = 95/5 drives the electron density to such low values in the core of the microdischarge during the pulse that there is not an immediate second avalanche in the core at the end of the pulse.

- After the pulse, the electron density shell propagates towards the center of the microdischarge due to high gap voltages in the core.

\[ RADIUS \text{ (µm)} \]

\[ \begin{align*}
\text{During Voltage Pulse} & : 1.1, 10.7, 20.4, 36.0 \\
\text{After Voltage Pulse} & : 40.1, 44.6, 67.8, 55.1, 40.7, 41.3
\end{align*} \]

\[ \text{Electron Density} \quad (10^{13} \text{ cm}^{-3}) \]

\[ \begin{align*}
\text{During Voltage Pulse} & : 0.5, 1.1, 2.0, 4.0 \\
\text{After Voltage Pulse} & : 40.1, 44.6, 67.8, 55.1, 40.7, 41.3
\end{align*} \]

\[ \text{Gap Voltage (kV)} \]

\[ \begin{align*}
\text{During Voltage Pulse} & : 0.5, 1.1, 2.0, 4.0 \\
\text{After Voltage Pulse} & : 40.1, 44.6, 67.8, 55.1, 40.7, 41.3
\end{align*} \]

- Xe/Cl₂ = 95/5,
  - Pressure: 0.6 atm,
  - Gas Temperature: 300 K,
  - \( V_o = 10 \text{ kV} \),
  - \( \tau = 40 \text{ ns} \),
  - \( \varepsilon = 5 \varepsilon_0 \)

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XeCl* AND 308 nm GENERATION IN Xe/Cl₂ = 95/5

- XeCl* in the core of the microdischarge is regenerated as the second avalanche moves towards the center after the voltage pulse.
- The total number of 308 nm photons is similar to the case of Xe/Cl = 99/1.

- 1 atm, 400 K, $V_o = 12$ kV, $\tau = 40$ ns, $\varepsilon = 5$
OTHER PHOTONS IN Xe/Cl$_2$ = 95/5

- For the 5% Cl$_2$ gas mixture, the generation of 172 nm and 490 photons dramatically decreases and the generation of 259 nm photons increases, compared to Xe/Cl$_2$ = 99/1.

- This trend is due to the fact that Cl$_2$ is very efficient at quenching Xe$_2^*$ and Xe$_2$Cl$_1^*$, and the higher Cl$_2$ mole fraction is beneficial to generating Cl$_2^*$.

- Xe/Cl$_2$ = 95/5, 0.6 atm, 300 K, $V_o = 10$ kV, $\tau = 40$ ns, $\varepsilon = 5$
• With increasing dielectric capacitance, the energy deposition and generated photons linearly increase.

![Graph 1: Energy Deposition for One Microdischarge (x10^-6 J)](image)

![Graph 2: Total Generated 308 nm Photons Per Unit Gas Volume in Microdischarge Region (x10^14 cm^-3)](image)
• With increasing dielectric capacitance, the efficiencies of photon generation decrease.

• This is because large dielectric capacitance leads to high plasma density which has a stronger quenching effect.
The efficiency of XeCl* 308 nm photon generation is influenced largely by gas pressure and slightly by Cl2 mole fraction and applied voltage.

\[ T_{\text{gas}} = 300 \text{ K}, \quad \tau = 40 \text{ ns}, \quad \varepsilon = 5 \]
DOE ANALYSIS: EFFICIENCY OF Xe$^*$ 172 nm PHOTON GENERATION

- The efficiency of Xe$^*$ 172 nm photon generation is influenced largely by Cl$_2$ mole fraction and slightly by gas pressure ratio and applied voltage.

Efficiency of 172 nm photon generation (%)

- Pressure = 0.6 atmosphere
- Applied Voltage = 10 kV
- Cl$_2$ = 3%

T$_{\text{gas}}$ = 300 K, $\tau$ = 40 ns, $\varepsilon$ = 5
DOE ANALYSIS: EFFICIENCY OF Xe₂Cl² 490 nm PHOTON GENERATION

- The efficiency of Xe₂Cl² 490 nm photon generation is influenced largely by Cl₂ mole fraction and slightly by gas pressure ratio and applied voltage.

\[ T_{\text{gas}} = 300 \text{ K}, \quad \tau = 40 \text{ ns}, \quad \varepsilon = 5 \]
DOE ANALYSIS: EFFICIENCY OF $\text{Cl}_2^*$ 259 nm PHOTON GENERATION

- The efficiency of $\text{Cl}_2^*$ 259 nm photon generation is influenced largely by $\text{Cl}_2$ mole fraction and slightly by gas pressure ratio and applied voltage.

$T_{\text{gas}} = 300 \text{ K}, \quad \tau = 40 \text{ ns}, \quad \varepsilon = 5$
CONCLUDING REMARKS

- Microdischarge dynamics in dielectric discharge have been investigated for high intensity sources of UV and VUV radiation emitted by dimer and trimer excimers.

- In Xe/Cl$_2$ gas mixture, the electron density show a shell expansion during the pulse due to dielectric charging and attachment of Cl$_2$ at small radii at lower E/N.

- The strong attachment of Xe/Cl$_2$ = 95/5 leads to shell propagation to smaller radii after the pulse.

- Increasing capacitance of the dielectric increases the energy deposition and the total light generation with a slight loss in efficiency.

- The efficiency of 308 nm photon generation is influenced mainly by gas pressure, decreasing with increasing pressure, and the efficiencies of 172, 259 and 490 nm photon generation are most sensitive to Cl$_2$ density.