Plasma assisted high pressure combustion; surface HP nanosecond DBD

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Plan of the presentation

Introduction: physics and chemistry of plasma-assisted combustion

Shock tube experiments: 0D, low pressures, high temperatures

Rapid compression machine (RCM) experiments: 2D, high pressures, low temperatures

High pressure high temperature (HPHT) discharge cell experiments: the discharge and the following combustion

High pressure discharge: streamer-to-filament transition
I. Physics and chemistry of plasma assisted combustion
Plasma assisted ignition/combustion: nonequilibrium plasma applications

- Lean mixtures
- Fast flows (1998)
- High pressures
Combustion: chain reactions

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Equation</th>
<th>Energy (kJ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initiation</td>
<td>$H_2 + O_2 = 2\text{OH} + 78\text{kJ}$</td>
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</tbody>
</table>
| Branching       | $\dot{H} + O_2 = \text{OH} + \dot{O} + 70\text{kJ}$  
                 | $\dot{O} + H_2 = \text{OH} + \dot{H} + 8\text{kJ}$ |             |
| Development: 2x | $\text{OH} + H_2 = H_2O + \dot{H} - 62\text{kJ}$ |             |
| Break           | $\dot{H} + \text{wall}$  
                 | $\dot{H} + O_2 + M = \text{HO}_2 + M - 203\text{kJ}$ |             |
Comparison of the reaction rates

\[ e + \text{O}_2 = e + \text{O} + \text{O} \]

\[ \text{O}_2 + \text{M} = \text{O} + \text{O} + \text{M} \]

\[ \text{H} + \text{O}_2 = \text{OH} + \text{O} \]

Constant rate, cm$^3$/s vs Temperature, K
Fast gas heating: time less than VT-relaxation

\[ k = B T^n \exp\left(-\frac{E_a}{kT}\right) \]
First available experiments on plasma-assisted combustion

Semenov N N (1958) Some problems of chemical kinetics and reactivity *Pergamon Press*

G. Gorchakov, F. Lavrov, Influence of electric discharge on the region of spontaneous ignition in the mixture $2H_2-O_2$. *Acta Physicochim. URSS* (1934), 1, 139-144
Energy branching vs reduced electric field $E/N$

Applied high voltage pulses

Positive and negative polarity pulses

Single pulse regime, 20 ns pulse duration, 2 ns front rise time
Kinetic approach to description of a gas discharge

- Current, voltage on electrodes
- Mixture composition
- Energy contribution
- Electric field \( E(\vec{r}, t) \)
- Electron density \( n_e(\vec{r}, t) \)
- Electron energy distribution \( f(\vec{r}, t, \varepsilon) \)
- Excitation of internal degrees of freedom of the gas
Field of parameters where nanosecond PAI/PAC experiments are available

All experiments were performed in a SINGLE-SHOT regime
II. Shock tube experiments: [relatively] low P and high T
Shock tube setup for plasma ignition

- Mixture composition
  \[ \text{CH}_4/\text{C}_2\text{H}_6/\text{C}_3\text{H}_8/\text{C}_4\text{H}_{10}/\text{C}_5\text{H}_{12} \]
  \(- \text{O}_2 – \text{Ar (90\%)}\)

- Temperature
  950-2000 K

- Pressure
  0.2-1.0 atm

- \(T_5, P_5\)
- \(T_{\text{ign}}\)
- \(E/N, I, W\)
Dielectric section of a shock tube

CH$_3$O; N$_2$; Ar; U=110 kV
$T_s=1300$ K, $P_s=0.5$ atm

Starikovskaia S M, Kukaev E N, Kuksin A Yu, Nudnova M M and Starikovskii
Comb. and Flame, 2004, 139, 177-87
The idea of the experiment

Ignition delay time (induction time)

Auto-ignition

O, OH, H, CH₃

Plasma assisted ignition
Shift of the ignition delay time: 
(CH₄:O₂):Ar = 90:10 mixture

Dissociation in a nanosecond discharge (C\textsubscript{2}H\textsubscript{6}:O\textsubscript{2}):Ar, Hayashi (C\textsubscript{2}H\textsubscript{6}), Braginsky (O\textsubscript{2}), Tachibana (Ar)

\[ T_5 = 1300 \text{ K}, \ P_5 = 0.65 \text{ bar}, \ w = 23.1 \text{ mJ/cm}^3 \]

\[ \text{Density, } 10^{15} \text{cm}^{-3} \]

\[ \text{E/n, Td} \]

Decrease of ignition delay time: moderate gas densities; uniform ns discharge

Kinetics of the ignition: kinetic curves \((T_5 = 1530 \text{ K}, n_5 = 5 \times 10^{18} \text{ cm}^{-3})\)

Plasma assisted ignition is characterized by:
- slow increase of gas temperature
- developed kinetics of intermediates
- partial fuel conversion during induction time

II. Rapid compression machine (RCM) experiments: high P and [relatively] low T
Rapid compression machine (RCM)
Nanosecond surface dielectric barrier discharge (nSDBD, for flow control)

Y Zhu, S Shcherbanev, B. Baron and S. Starikovskaia
*PSST, 26 (2017) 125004*
Cylindrical electrode system (for PAC)

Electrode configuration

Cubic HP chamber

- High-voltage electrode
- Dielectric material
- Grounded electrode
- Cable
- 0.3 mm PVC film
- Frontal view
- Side view
- HV electrode, 2 cm in diameter

To HV generator

Gas inlet
Rapid compression machine (RCM), Lille
Mixtures used in experiments:

1) CH$_4$/O$_2$/Ar, $\phi=1$, 0.5, 0.3, 70-75 % of Ar

2) n-C$_4$H$_{10}$/O$_2$/Ar/N$_2$, $\phi=1$, 38 % of N$_2$, 38 % of Ar

3) n-C$_4$H$_{10}$/O$_2$/Ar, $\phi=1$, 76 % of Ar

4) n-C$_4$H$_{10}$/O$_2$/N$_2$, $\phi=1$, 76 % of N$_2$
P-T diagram for RCM experiments

- (CH$_4$:O$_2$, $\phi=1$) + 72% Ar
- (CH$_4$:O$_2$, $\phi=0.5$) + 75% Ar
- (CH$_4$:O$_2$, $\phi=0.3$) + 77% Ar
- (n-C$_4$H$_{10}$:O$_2$, $\phi=1$) + 77% N$_2$
- (n-C$_7$H$_{16}$:O$_2$, $\phi=1$) + 79% N$_2$

- n-C$_4$H$_{10}$
  (cool flame modification)
- n-C$_7$H$_{16}$

Pressure $P_{TDC}$ / bar vs. Temperature $T_c$ / K

No autoignition
Autoignition vs plasma ignition in RCM at $P_{TDC}=15$ bar and $T_C=970$ K, $(CH_4:O_2)+76\%$Ar
Pressure trace and corresponding fast imaging of flame propagation


$\text{CH}_4:O_2, \text{ER}=1 + 70\% \text{Ar}$, $T_C=947 \text{ K}$, $P_{TDC}=15.4 \text{ bar}$
Ignition threshold and polarity of the high-voltage pulse

limit of filamentation in air

- n-C$_4$H$_{10}$/O$_2$/N$_2$, U>0
- n-C$_4$H$_{10}$/O$_2}$/Ar, U>0
- n-C$_4$H$_{10}$/O$_2}$/N$_2$, U<0
- n-C$_4$H$_{10}$/O$_2}$/Ar, U<0

Threshold voltage, kV vs. P/T, bar/K
III. High pressure high temperature (HPHT) chamber experiments:
high P and low T [T=300 K]
High-Pressure and High-Temperature (HPHT) discharge/combustion chamber

Electrode configuration

- Quartz
- Ceramics
- HV Generator
- Grounded electrode
- High-voltage electrode

General view of the HPHT setup

Discharge/combustion chamber
Experimental setup

The scheme of experimental setup. SR – Spectrograph, ICCD – camera, PC – computer, BCS – back current shunt,
nSDBD and Flame Initiation, P=3 bar, U=-50 kV

Discharge (ns) and combustion (ms) emission patterns

Discharge, SPS

Flame, OH

ns discharge

Intermediate chemistry

Flame propagation, Combustion

\[ \text{H}_2: \text{Air, ER}=0.6 \]

\[ \text{Intensity, a.u.} \]

\[ \text{Wavelength, nm} \]

\[ \text{Intersity, a.u.} \]

\[ \text{Wavelength, nm} \]

\[ \text{OH} \]

\[ \text{SPS} \]

\[ \text{O}, \text{OH}, \text{H}, \text{HO}_2, \text{H}_2\text{O}_2, \text{H}_2\text{O}, \text{etc.} \]

\[ \text{N}_2(\nu), \text{N}_2(\text{A,B,C,...}), \text{H}, \text{O}(^1\text{D}), \text{O}_2(\text{a}^1\Delta_y), \text{etc.} \]
Initiation of Combustion with nSDBD

H₂/Air mixture, P=3 bar, T₀=300 K, ER=0.6

PM signal, V

- Yes, for combustion!
- No, for plasma chemistry

Discharge
Pre-flame
Ignition delay?
Ignition

Delay: ICCD gate: 50 µs

50 µs
100 µs
200 µs
400 µs
Flame Initiation in H₂/Air ER=0.5, P=6 bar

First regime of ignition:
Ignition kernels

Polarity: U>0
Energy deposition
W= 3 mJ

Ignition with a few ignition kernels near HV electrode. Streamer discharge. Pressure 6 bar, Temperature 300 K.
Flame Initiation in $\text{H}_2$/Air ER=0.5, P=6 bar

Second regime of ignition:
Ignition along the perimeter of HV electrode

Polarity: $U>0$
Energy deposition $W=4.8$ mJ

Quasiuniform ignition around HV electrode. Streamer discharge.
Pressure 6 bar, Temperature 300 K.
Flame Initiation in H₂/Air ER=0.5, P=6 bar

Third regime of ignition:
Ignition along the discharge channels

Polarity: U>0
Energy deposition
W= 12 mJ

Ignition along the channels. Filamentary discharge.
Pressure 6 bar, Temperature 300 K.
Discharges in different gas mixtures

Discharge in methane/O$_2$/Ar

Discharge in n-heptane/O$_2$/Ar

Discharges in CH$_4$/O$_2$/Ar (ER=0.6) and n-C$_7$H$_{16}$/O$_2$/Ar (ER=1) mixtures, $P_0=3$ bar, $T_0=300$ K, voltage on the electrode $U=+38$ kV
IV. High pressure surface DBD discharge: streamer-to-filament transition
Two modes of nSDBD (velocity is a few mm/ns)

Electrode system

Filamentary mode, $V=\text{-}46 \text{kV}$, 4 bar, Air

Cylindrical electrode configuration
Outer diameter 50 mm; HV electrode diameter 20 mm

D=2 cm HV electrode

HV electrode

Camera gate is 0.5 ns
Analysis of emission intensity (Streamer vs filamentary mode)

Radial distribution of the frontal discharge emission; \( P=4 \) bar, \( U=-47 \) kV.

\[
\frac{I_{11\text{ns}}^{\text{fil}}}{I_{11\text{ns}}^{\text{dif}}/I_{4\text{ns}}^{\text{dif}}} \approx 50/1.5/1
\]
Spatial distribution of emission

<table>
<thead>
<tr>
<th></th>
<th>No filter</th>
<th>Filter (340±5) nm</th>
<th>Filter (532±5) nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Streamers</td>
<td><img src="image1.png" alt="Image" /></td>
<td><img src="image2.png" alt="Image" /></td>
<td><img src="image3.png" alt="Image" /></td>
</tr>
<tr>
<td>Filaments</td>
<td><img src="image4.png" alt="Image" /></td>
<td><img src="image5.png" alt="Image" /></td>
<td><img src="image6.png" alt="Image" /></td>
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</tbody>
</table>
How to get the filament spectra

Single filament is aligned with the entrance slit of the spectrometer
Continuous spectra in filamentary discharge
Broadening of \( H_\alpha \) line

Filamentary discharge in \( \text{N}_2: \text{H}_2 \) (7:1) mixture \( P = 8 \) bar. Electron density measurements with \( H_\alpha \) line (656 nm) broadening. Camera gate 5 ns.
Recombination emission in the filament

Electron density from recombination emission

SPS (337.1 nm) is a reference radiation

\[
\begin{align*}
\text{N}_2(\text{C}) &= k_c(E/N) \cdot n_e \cdot \text{N}_2/ \nu_q \\
Q_c &= \text{N}_2(\text{C}) \cdot F_{FK} A_{00} \\
Q_c &= 3 \cdot 10^{21} \text{quantum/cm}^3/\text{s}.
\end{align*}
\]

\[Q_C(t=0 \text{ ns})/Q_{CW}(t=5 \text{ ns}) \sim (1.5-2)\]

\[Q_{CW}(\omega) = C_0 \cdot \frac{n_e \cdot n_{\text{ion}}}{\sqrt{T_e \cdot \eta \omega}} d\omega = (1.5 - 2) \cdot 10^{21} \text{quantum/cm}^3/\text{s}\]

At \( \lambda = 337 \text{ nm} \) and \( \Delta \lambda = 3 \text{ nm} \), \( d\omega = 5 \cdot 10^{13} \text{ s}^{-1} \)

In the discharge \( T_e \sim 2-4 \text{ eV} \) \( \rightarrow n_e \sim 3 \cdot 10^{18} \text{ cm}^{-3} \)
In the afterglow \( T_e \sim 0.5 \text{ eV} \) \( \rightarrow n_e \sim 10^{18} \text{ cm}^{-3} \)
Conclusions

Nanosecond dielectric barrier discharge (nSDBD) provides ignition of different fuels in a wide range of stoichiometries.

At high gas densities, nSDBD simultaneously provides hundreds of ignition sites distributed in space. The geometry of the ignition and following combustion can be adapted to provide the highest possible efficiency of the system.

Physics of filamentation in high pressure nSDBD is under study now ($n_e=10^{19}$ cm$^{-3}$ is achieved during a few nanoseconds in a single-shot mode).