Nanosecond discharges: high power density for distributed in space ignition

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Outline of talk

**Introduction.** High pressures (7-15 bar), low T (600-1000 K). Do we need very high [O] densities for plasma assisted ignition (PAI)?

**Rapid compression machine (RCM):** ignition by nanosecond discharge. Analysis of experimental results and perspectives

**Surface dielectric barrier discharge (SDBD):** quasi-uniform and filamentary mode

**Surface dielectric barrier discharge (SDBD):** what can we get from E-field measurements by emission?

**Conclusions**
A typical ignition length for uniform flow

- **150 ms** × **2.5 m/ms** (M=8) = **375 m**

- **TGV Eurostar**: TransManche Super Train, **393.72 m** (20 cars)
History of plasma assisted combustion

- **1998 to 2001**: Initial research and development
- **2004**: Significant advancements in plasma technology
- **2007**: Integration of plasma-assisted combustion in power plants
- **2010**: Increasing use in industrial processes
- **2013**: Continued innovation and expansion of applications

Key Diagram Elements:
- **Overview of Plasma Chamber**: Depicts the components and flow of plasma-assisted combustion.
- **Airflow M-2**: Illustrates the initial stages of airflow and injection.
- **Propane Injection**: Highlights the role of fuel in plasma combustion.
- **OH and CH^* Emission**: Graphs showing emission levels with and without plasma discharge.
- **Measuring Emission**: Diagram for tracking the emission of plasma discharge benefits.
Available reviews

Starikovskaia (J. Phys. D 2006)

Popov (High Temp. 2007)

Adamovich (PSST, 2009)

Starikovskaia and Starikovskiy (In: Handbook of Combustion, 2010)

Starikovskiy and Aleksandrov (Aeronautics & Astronautics 2011)


Starikovskiy and Aleksandrov (Progr. Energy Comb. Sci. 2013)
Close of faraway from the combustion threshold: what is the difference?

I: wall recombination

II: volume recombination

III: heat dissipation


Parameters of Shock Tube PAI experiments: temperature, pressure, ignition delay time

T₅, K

Number of C atoms

1 2 3 4 5

1200 1400 1600 1800 2000

PAI

Autoignition

n₅, 10⁻¹⁸ cm⁻³

Number of C atoms

1 2 3 4 5

0 2 4 6 8

PAI

Autoignition

Ignition delay time, µs

10⁻¹ 10⁰ 10¹ 10² 10³

10-30 mJ/cm³

Number of C atoms

1 2 3 4 5

NeQ MIPT Lab, Moscow, 1998-2008
Active species produced by plasmas and their role in kinetics

- Rotationally excited molecules (RT-relaxation)

- Vibrationally excited molecules (VT-relaxation + acceleration of chain branching/prolongation)

- Electronic states (dissociation, chemistry, \( \Delta T \))

- Atoms and radicals (chemistry, \( \Delta T \))

- Charged particles (chemistry, heating)

\[ k = A \exp(-\frac{E_a}{RT}) \]
High pressures, low temperatures

Pressure: 10-15 bar

Temperature: 600-1000 K
Induction time as a function of “atomic oxygen density” (difference: discharge & cool flame)

\((C_{4}H_{10}:O_{2}, \text{ER}=1):\text{Ar}, P=8.6 \text{ bar}, T=800 \text{ K}\)

Artificial action: \(O_{2} \rightarrow O + O\)

Remark: why discharge is NOT a cool flame

Butane stoichiometric mixture diluted by 76% of Ar

$P_0 = 8.6$ bar
$T_0 = 800$ K

Ignition

Equilibrium: chemistry leads to $\Delta T$

Relaxation of $W$ leads to $\Delta T$ and initiates chemistry
Ignition delay at additions of O-atoms: weak dependence at [O]>0.5%

Butane stoichiometric mixture diluted by 76% of Ar

PTDC=8.6 bar
Tc=800 K

Percent of initially dissociated O₂

Induction delay time, ms
Energy cost for active species in C$_2$H$_6$:O$_2$ mixture: too much dissociation?

Energy cost, eV/particle

$O_2(a^1\Delta_g)$, $CH_3$, $O(^3P)$, $H$

Energy $W=50$ mJ; volume $V=5$ mm$^3$: [$O$] is about $10^{19}$ cm$^{-3}$

PSST, 21(2012) 045012
ICCD image (camera gate is 0.5 ns) of discharge in 1 atm air: “combined” SDBD
All the experiments are performed in SINGLE-SHOT regime
Electrode system and applied pulses

±(25-50) kV pulses, 15-25 ns duration, 0.5-3 ns rise time
Machine at PC2A laboratory, Lille University
Combustion chamber for RCM with discharge
Initial parameters of the experiments

Temperature and pressure

RCM core temperature: $T_C = 640-980$ K

Pressure at Top Dead Center: $P_{TDC} = 7.5-16$ atm

High-voltage pulse

Pulse duration: 25 ns, front rise time: 0.5 ns

Pulse amplitude on the electrode: $\pm (24-54) \text{ kV}$
Mixtures used in the high pressure (RCM) experiments

1) $\text{CH}_4/\text{O}_2/\text{Ar}$, $\phi=1$, 0.5, 0.3, 76 % of Ar

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

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

4) $\text{C}_4\text{H}_{10}/\text{O}_2/\text{N}_2$, $\phi=1$, 76 % of N$_2$
Methane is not the easiest fuel to burn

\[
\text{Comb. & Flame, 2008, 2009}
\]
Aldehydes and chain branching:

\[ RO_2 \rightarrow R' + R''CHO \]

\[ R''CHO + O_2 \rightarrow R''CO + HO_2 \]

(P, T) - field of RCM ignition experiments

Region of calculations

- **No autoignition**
- **Region of calculations**

- (CH₄:O₂, φ=0.5) + 75% Ar
- (CH₄:O₂, φ=0.3) + 75% Ar
- (C₄H₁₀:O₂, φ=1) + 38% Ar + 38% N₂
- (C₄H₁₀:O₂, φ=1) + 76% N₂
- (C₄H₁₀:O₂, φ=1) + 76% Ar
- (CH₄:O₂, φ=1) + 76% Ar

**Pressure P_d, atm**
**Temperature T_c, K**

n=2 n_{atm}

n=4.3 n_{atm}
Auto- and plasma assisted ignition. CH$_4$:O$_2$:Ar, ER=1, 71% of Ar

Induction time decreases significantly
Possible knocking suppression (CH₄:O₂:N₂:Ar, T=730 K)

Confirmed statistically (30 experiments)
Cool flame regime modification under the discharge action

Mixture: $\text{C}_4\text{H}_{10}/\text{O}_2/\text{N}_2$, $\phi = 1$, $T_c = 710$ K

Intermediate modification of pressure profile occurs in experiments with cool flames
Definition of the ignition delay time

Pressure, atm

Time, ms

Discharge initiation

\[ \tau \]

\[ \frac{P}{2} \]

\[ P \]
Ignition delay time vs voltage/deposited energy. CH$_4$O$_2$:Ar, ER=0.3 and 0.5, 76% of Ar

- CH$_4$/O$_2$/Ar, φ = 0.3, 76% of Ar
  - PTDC = 15.5 atm
  - TC = 962 K

- CH$_4$/O$_2$/Ar, φ = 0.5, 76% of Ar
  - PTDC = 15.1 atm
  - TC = 943 K

ER = 0.3

ER = 0.5
Ignition delay time vs deposited energy. 
CH$_4$\text{:O$_2$\text{:Ar, ER}=0.3, 76\% of Ar

\[P_{TDC}=15.1 \text{ atm} \]
\[T_c=943 \text{ K} \]
MIE: minimal ignition energy, CH₄:O₂:Ar

ns PAI: 15 bar/960 K

MIE: 1 bar/300 K

![Graph showing the relationship between MIE (minimal ignition energy) and the percentage of CH₄ in the mixture. The graph indicates a curve where MIE decreases as the percentage of CH₄ increases up to a certain point, after which MIE increases again.]

- MIE, 1 bar, 300 K
- PAI-total, 15 bar, 960 K
- PAI/channel (qu), 15 bar, 960 K
- PAI/channel (fil), 15 bar, 960 K

MIE, mJ

% of CH₄ in the mixture

<ms
Suppression of NTC phenomena by nanosecond discharge (10-20 mJ)
NTC region calculation: P=8.6 bar, T=800 K

Buthane stoichiometric mixture diluted by 76 % of Ar

Experimental results:
- T=822 K, P=(8.7-7.8) bar, $\tau=65$ ms
- T=844 K, P=(8.6-7.7) bar, $\tau=68$ ms
- T=843 K, P=(8.6-7.7) bar, $\tau=68$ ms

Experiments are in this region
Calculated (smooth) change of ignition delay time

Butane stoichiometric mixture diluted by 76 % of Ar

$P_0 = 8.6$ bar
$T_0 = 800$ K

OH fraction

Time, ms

Ignition

Cool flame

0.5% of O

0.05% of O
High speed flame propagation imaging.
LaVision Phantom v9 camera

\( \text{CH}_4/\text{O}_2/\text{Ar}, \, \Phi = 1, \, 76\% \, \text{Ar}, \, T_C = 911 \, \text{K}, \, P_{TDC} = 15.4 \, \text{bar} \)
Induction time as a function of deposited energy (Popov’s mechanism)

Methane/oxygen mixture diluted by 76 % of Ar

\[ \text{Deposited energy, eV/mol} \]

\[ \text{Induction time, ms} \]

Heating and production of O-atoms are taken into account

\[ P_{\text{gas}} = 960 \text{ K} \]
\[ P_{\text{gas}} = 15 \text{ bar} \]

14 mm
Preliminary modeling: coupling of afterglow-combustion kinetics?

Density, cm\(^{-3}\)

Time, ns

- O\(^{3}\text{P}\)
- CH\(_3\)
- CH\(_2\)O
- H\(_2\)O
- H
- OH
- O\(^{1}\text{D}\)

0.05 eV/mol
- 24 kV discharge; O$_2$: Ar; T=300 K; P=3.2 bar

U = + 24 kV on the electrode

2 ns ICCD gate
+ 24 kV discharge; O₂: Ar; T=300 K; P=3.2 bar

U = + 24 kV on the electrode
ICCD imaging: air, gate=2 ns. Emission of the $2^+$ system of molecular nitrogen

Quasi-uniform ("diffusive") and filamentous modes

P=3 atm

\[ U = -35 \text{ kV} \]

P=5 atm

\[ U = -55 \text{ kV} \]
Oscillograms corresponding to different high pressure discharge modes

3 atm, -35 kV

5 atm, -55 kV
Uniform-filamentary transition: 337 nm spectra

- Wavelength, nm
- Intensity, a.u.

**P=1 atm, U=-24 kV**
- t=5 ns

**Wavelength, nm**
- 335.0 335.5 336.0 336.5 337.0

**Intensity, a.u.**
- 0.01 0.1 1

- region 0-2 mm from HV electrode
- region 3-5 mm from HV electrode
- theoretical fit, T=360 K

**t=40 ns**

**Wavelength, nm**
- 335.0 335.5 336.0 336.5 337.0

**Intensity, a.u.**
- 0.01 0.1 1

- Filamentous
- Theoretical fit, T=420
Transition between quasi-uniform and filamentous modes, synthetic air

Voltage amplitude on HV electrode, kV vs. Pressure, bar

Negative polarity pulses
Transition between quasi-uniform and filamentous modes, synthetic air

- **P= 3 bar, U=-47 kV on the HV electrode**

Diffusive beginning (first 6 ns)  | Transition to filaments  | Filamentous

- **2 ns**
- **6 ns**
- **15 ns**
Uniform-filamentary transition, $P=3$ atm in synthetic air: deposited energy
Uniform-filamentary transition, P=3 atm, air: currents and emission

Electrical current

337 nm emission
Electric field in SDBD: emission measurements, 391/337 ratio

\[
\frac{k_B}{k_C} = \left[ \frac{N_2^+(B)}{N_2(C)} \right] \times \frac{1}{\tau_{0B}} + \frac{1}{\tau_{0C}} \left( \Sigma k_q^{N_2^+(B)}[M] + \Sigma k_q^{N_2^-(C)}[M] \right) \tag{**}
\]

\[
\left[ N^* \right] \equiv \frac{I_{lu} m_0}{h v_v v'' A_{v_v v''}} \quad \Rightarrow \quad \frac{k_B}{k_C} = \left( \frac{I_B}{I_C} \right) \times K \quad \text{Measured}
\]

\[
k_e x = \int \sigma(\varepsilon) \sqrt{\frac{2\varepsilon}{m_e}} \sqrt{\varepsilon} f(\varepsilon) d\varepsilon \quad \Rightarrow \quad \left( \frac{k_B}{k_C} \right) = f \left( \frac{E}{N} \right) \quad \text{Calculated}
\]
Emission measurements, 391/337 ratio: experimental calibration

$R_{391/337} = 0.07$

$E/N = 600$ Td

38 (2005) 3894–3899
Intensity of 391.4 and 337.1 nm emission; Electric field in air at 4 atm and +24 kV

- assumption about the plasma uniformity: not true
Comparison of “E-field” in positive and negative polarity pulses

Electric field is higher in case of positive polarity pulses

High values of the E-field right after peak
Comparison of “E-field” in positive and negative polarity pulses vs pressure

Discharge: $T=300$ K; $P=5$ atm

RCM: $T=900$ K; $P=15$ atm

$W=10-30$ mJ

- higher values of the electric field for positive polarity
- higher $P \rightarrow$ lower $E_{\text{max}}$
Principal explanation of observed results

\[ I_{\text{measured}} = I_{ch} + I_{\text{gap}} \]

The same order of \( I_{ch} \) and \( I_{\text{gap}} \) is possible.
Results of numerical modeling (Soloviev)

$E/N = f(x,y,t)$
$I(N_2(C)) = f(x,y,t)$
$I(N_2^+(B)) = f(x,y,t)$
$I(N_2(C))$ and $I(N_2^+(B))$ integrated over Y

Ratio of integrals
Electric field in the discharge: analysis of numerical modelling

U = -24 kV: 400-430 Td
U = +24 kV: <200 Td
Electric field in the discharge: analysis of numerical modelling

Calculations

-24 kV, t=2.15 ns
+24 kV, t=4.85 ns
+24 kV, t=0.75 ns
Electric field in the discharge: analysis of numerical modelling

![Graph showing ratio of integrals, A, and base curve, R(391/337), against E/N, Td. The graph compares measurements and calculations for +24 kV and -24 kV.]
Conclusions

Low T, high P: The ignition delay time decreases significantly under the action of nanosecond SDBD, from tens and hundreds milliseconds to units of milliseconds at deposited energy within a range 10-30 mJ.

It is possible to describe qualitatively the results taking into account O-atoms production and \( \Delta T \). Starting from some O-density, ignition delay time remains close to a fixed value determined by gas mixture composition.

NTC region and cool flame kinetics are significantly influenced by addition of radicals. Detailed discharge-combustion mechanism for heavy hydrocarbons is a challenge.

Uniform discharge is a challenge. Discharge study is necessary for each range of conditions. Multipoint ignition is efficient.
Acknowledgements

- EOARD AFOSR

- Russian Foundation for Basic Research


- PUF Collaborative Grant (Ecole Polytechnique-Princeton University)

- French Academy of Science (CNRS) / Russian Academy of Science (RAS) collaborative grant No 23994 2010-2012) and PICS-RFBR grant (5745-11.02.91063-a/5745)
Thank you for your attention…
…You are invited

4th Aerospace Thematic Workshop
“Fundamentals of aerodynamic-flow and combustion control by plasmas”
CNRS Conference Centre Paul Langevin, Aussois, France
7-12 April, 2013; the next is in 2015

Questions:
Restrictions for March 2013?
Program?
- The PAI/PAC problem can be considered in 0D;

- Plasma is nonequilibrium (\(T_e \gg T_g\), shorter than needed to arc transition) and can be modeled by \(E/N(t)\), [70-200 Td], and \(n_e(t)\);

- Initial mixture parameters are the following: (F+O+D) mixture of known composition;
- Initial gas pressure is P;
- Initial gas temperature is T
Plasma action (tens-hundreds of ns)

\[
\{E/N(t), n_e(t)\} \rightarrow \text{EEDF}(t) \rightarrow M^+, M^*, A^*, M(v)
\]

**Advantages:** For majority of oxidizers and diluters, the discharge action is well-known: the sets of cross-sections measured by using electron beams technology (60th-70th) are self-consistent and verified on the basis of the experimentally measured swarm parameters (ionization coefficient, drift velocity, electron mobility)

**Drawbacks:** For fuels, no self-consistent data set is available for higher hydrocarbons
Afterglow (hundreds of ns - ms)

\[ \{ n_e, M^+, M^*, A^*, M(v), E/N->0 \} \rightarrow \text{kinetics, } T \]

**Advantages:** "plasma" and "combustion" kinetics are separated in time; "plasma" kinetics with participation of electronically and vibrationally excited species is well known at room temperatures for \( N_2, O_2, Ar \ldots \)

**Drawbacks:** the main classes of reactions are well-known, but a lack of data is observed when trying to describe the details of minor species behavior in chemically active mixtures
Ignition: $\mu s$ – ms, $f(P, T, \text{composition})$

Advantages: main classes of plasma-gas interaction are well-known, there is no risk to miss a whole class of chemical transformation; although special attention and detailed literature analysis is needed to know the limits of used kinetic schemes.

Drawbacks: There is no ready mechanism for the low-temperature ignition: it must be built as an intersection of available “pure” oxidation mechanism, “common” combustion mechanism and “plasma” action.

$\{M^*, A^*, M(v), T\} \rightarrow \text{“combustion” kinetics}$
Peculiarities and universal (C1-C3) and extended (low T) mechanisms

\[ \text{CH}_4/\text{C}_2\text{H}_6/\text{C}_3\text{H}_8 \] 
oxidation at high P and high, intermediate and low T (2008)

289 species
1580 reactions

D. HEALY, H.J. CURRAN, J.M. SIMMIE et al, 
ICCD imaging: air, gate=0.5 ns, P=1 atm, U=-22 kV (time in ns)

Ignition by spark discharge:
1 atm of C\textsubscript{2}H\textsubscript{6}:O\textsubscript{2}=2:7, T\textsubscript{in}=300 K

Single pulse spark discharge ignition of C\textsubscript{2}H\textsubscript{6}:O\textsubscript{2}=2:7 at 1 bar and ambient temperature.

Plasma Sources Sci. Technol. 21 (2012) 045012 (15pp)
Ignition by surface DBD at 1 atm, C$_2$H$_6$:O$_2$
Preliminary numerical modelling: CH$_4$:O$_2$:Ar. P=20 bar; T=950 K

Autoignition

Ignition initiated by 0.05% of O

45 ms

9 ms
Preliminary numerical modelling: n-C$_4$H$_{10}$:O$_2$:Ar. P=8 bar; T=700 K

Autoignition: 43 ms

Cool flame: 37 ms

Ignition initiated by 0.05% of O: 6 ms

Cool flame: 2 ms
Comparison of experiments and numerical modeling: all (CH$_4$-C$_5$H$_{12}$) mixtures

Ignition delay time, $\mu$s

1000/T, K$^{-1}$

- Auto Exp, C$_2$H$_6$
- Auto Calc, C$_2$H$_6$
- PAI Exp, C$_2$H$_6$
- PAI Calc, C$_2$H$_6$
- C$_3$H$_8$
- C$_4$H$_{10}$
- C$_5$H$_{12}$

CH$_4$, auto, 0.4-0.7 atm

CH$_4$, auto, 2 atm

C$_2$H$_6$-C$_5$H$_{12}$, auto, 0.2-0.7 atm

PAI, 0.2-0.7 atm