

Fundamental Mechanisms, Predictive Modeling, and Novel Aerospace Applications of Plasma Assisted Combustion

**AFOSR
MURI Review Meeting**

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October 22, 2013

Report Documentation Page

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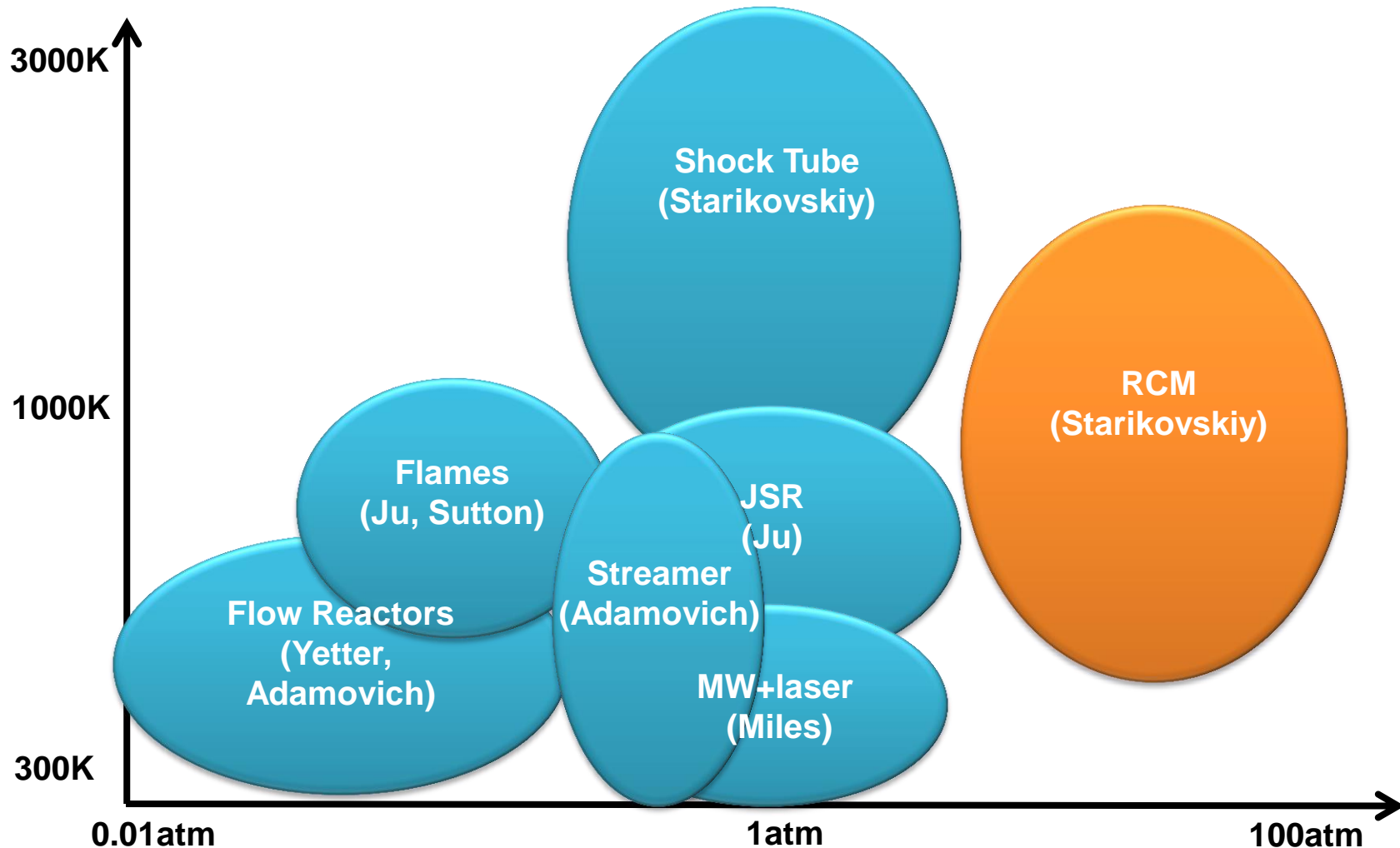
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Main Tasks

- **High Temperature**
- **High Pressure**
- **High Speed**
- **High Voltage**
- **High Power**

MURI Deliverables: Kinetic Data Generation

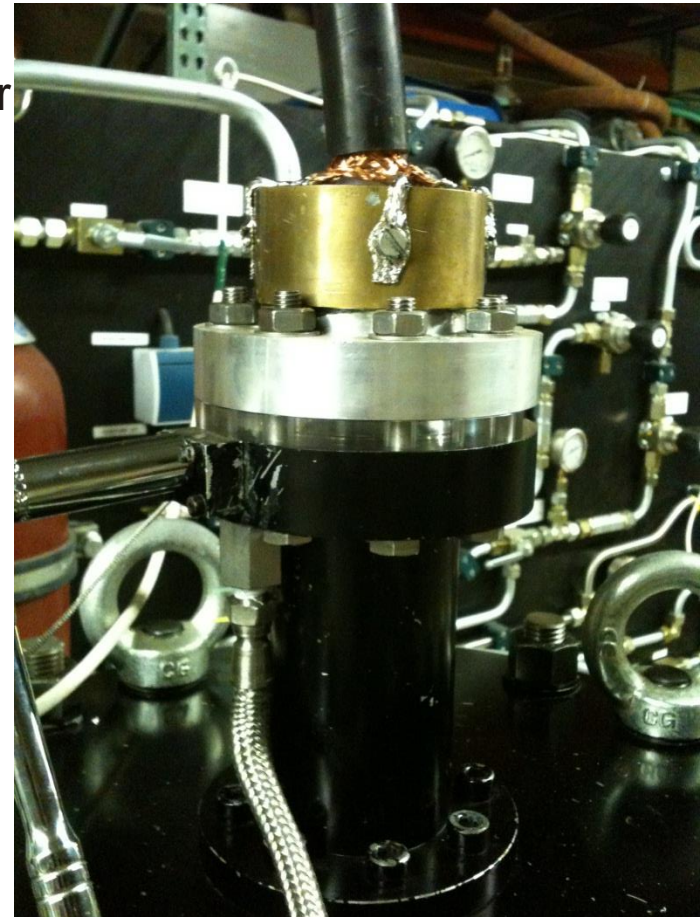
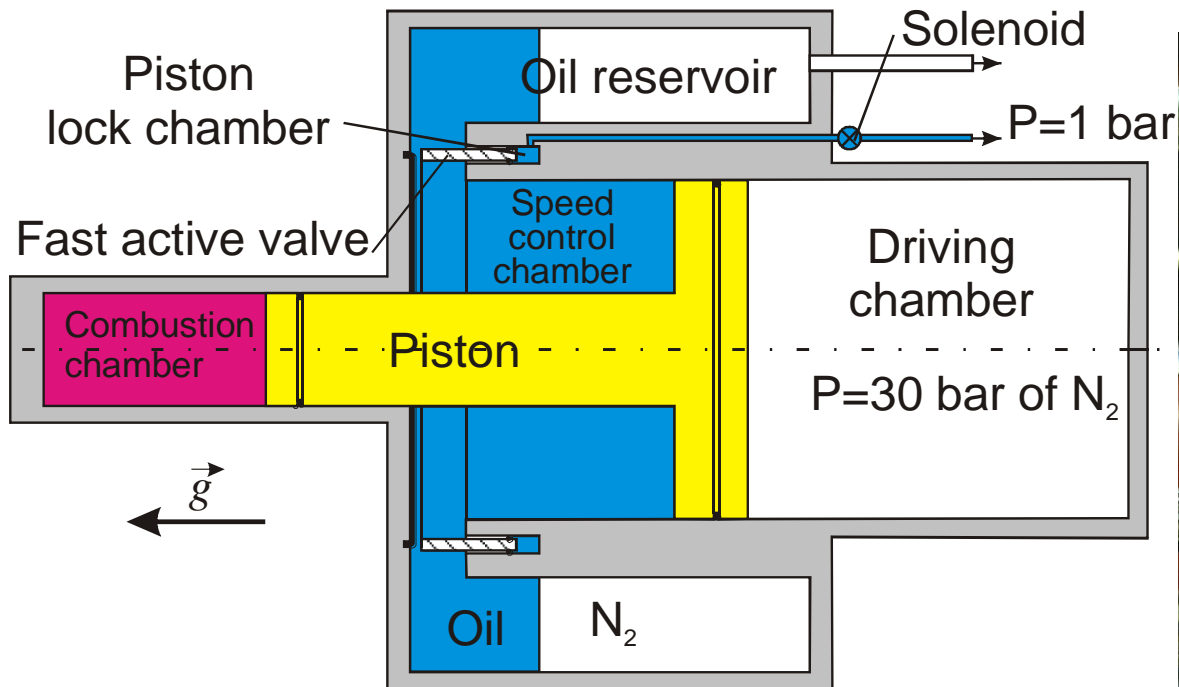


Rapid Compression Machine:

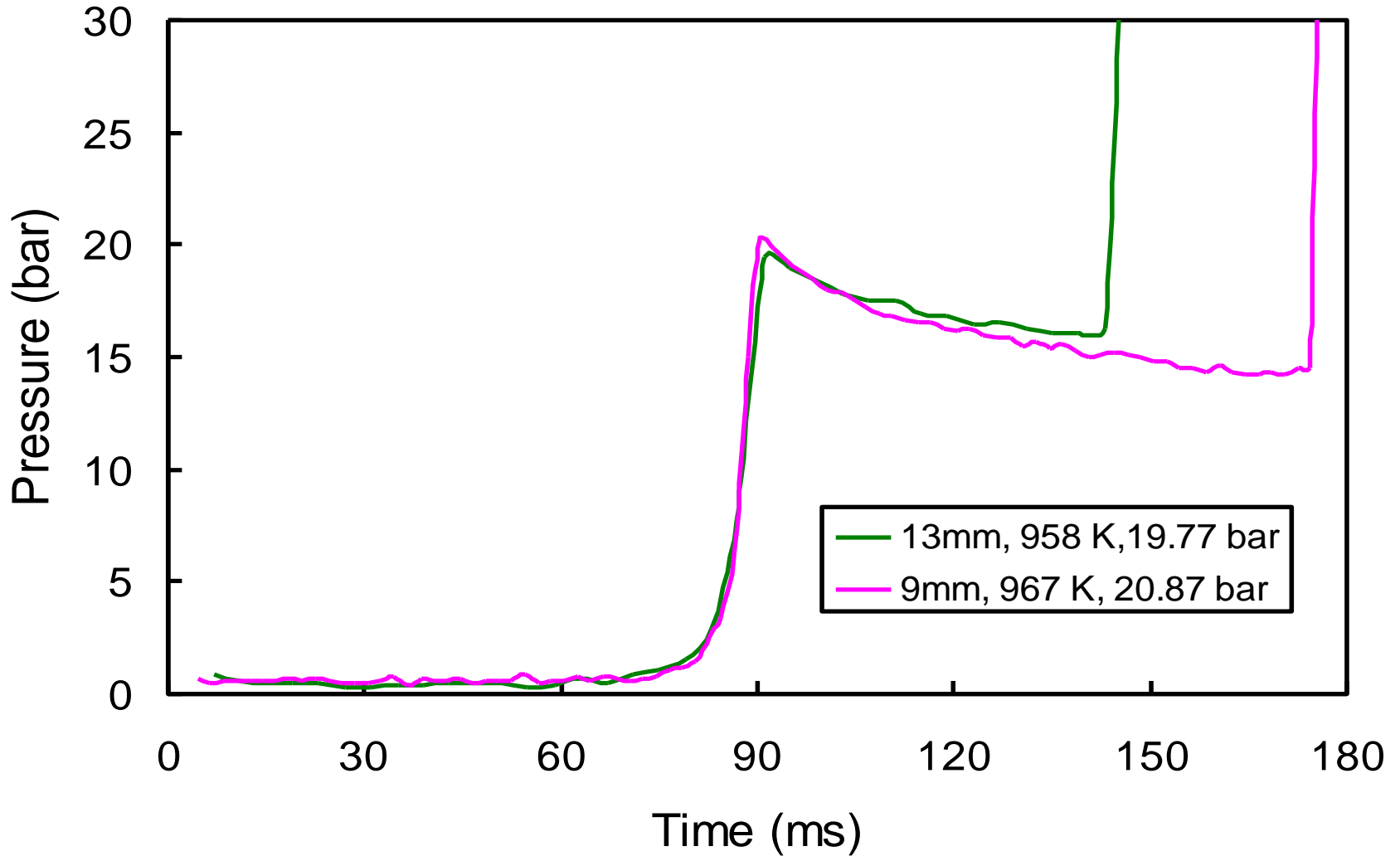
$P = 10\text{-}70 \text{ atm}$, $T = 650\text{-}1200 \text{ K}$



Scheme of the RCM

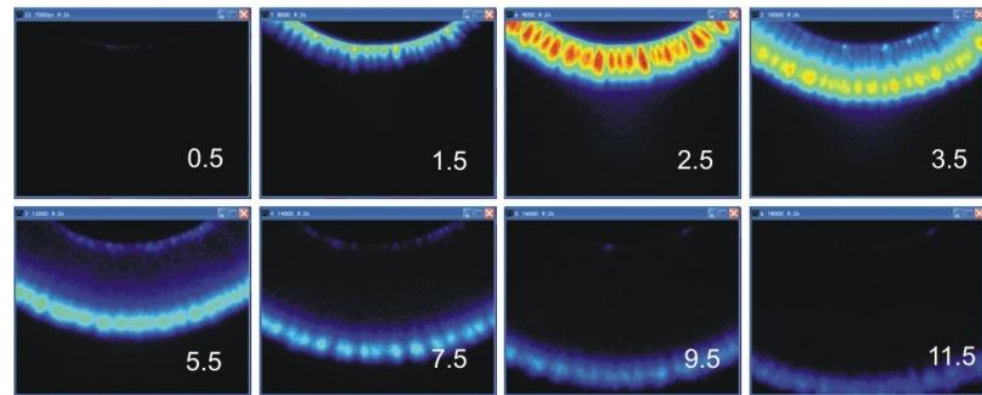


Gas Dynamic Limitations

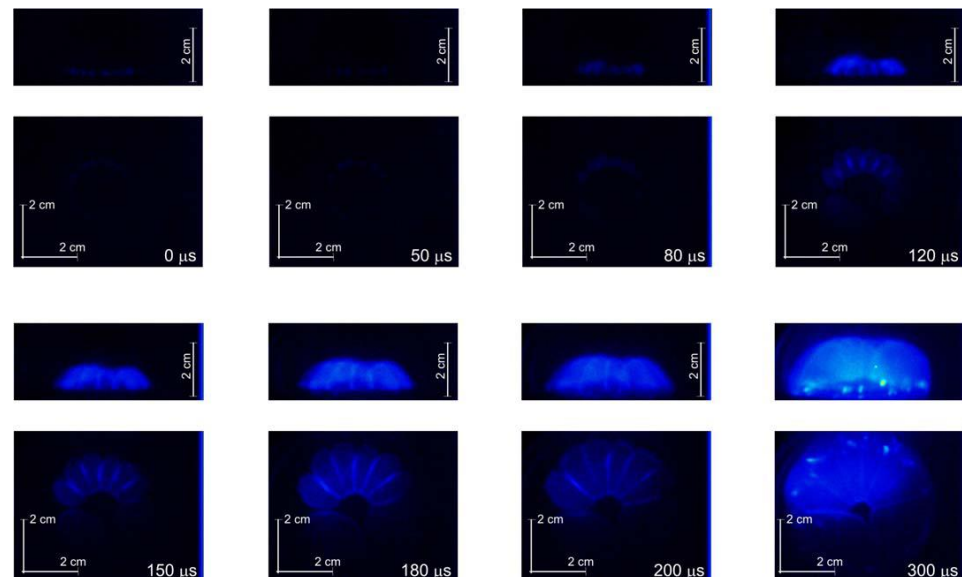


Gas Discharge Limitations

ICCD images of the discharge at 1 atm dry air. Negative polarity of the high-voltage electrode, 22 kV, 25 ns duration, $f = 40$ Hz [Kosarev et al, 2009].



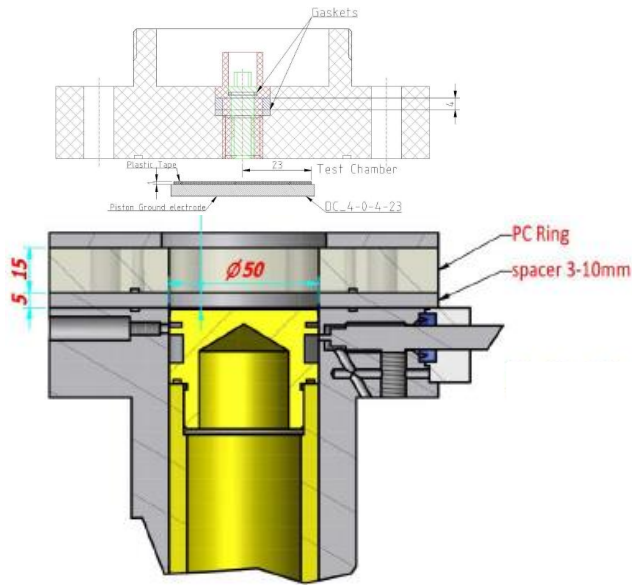
Mixture $C_2H_6:O_2=2:7$ at 1 bar and ambient initial temperature was successfully ignited in ~ 100 ms in relatively large volume [Sagulenko et al, 2009].



SDBD Development at High Pressures

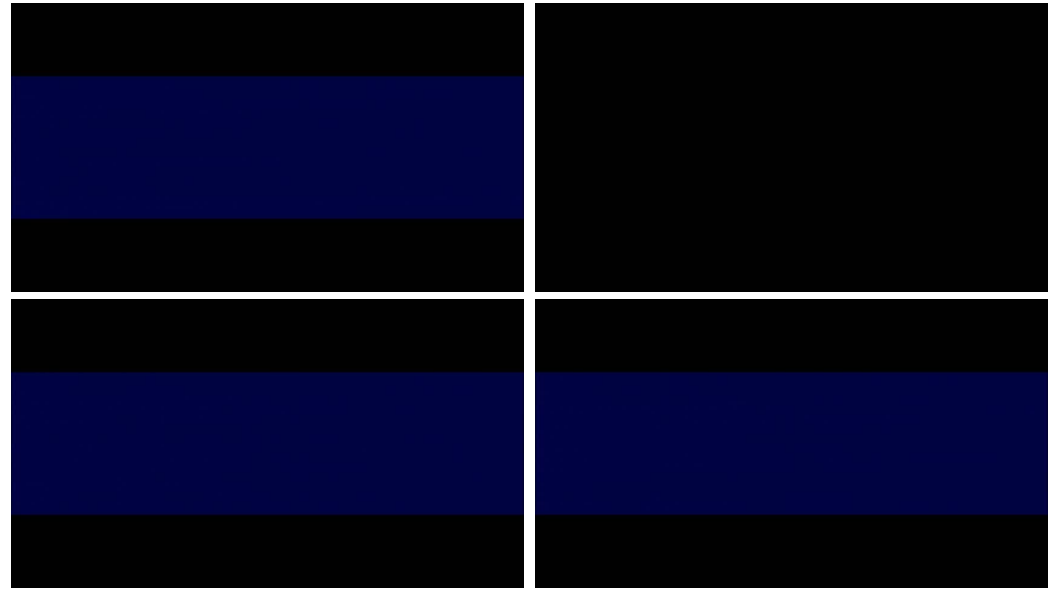
1 atm

4 atm



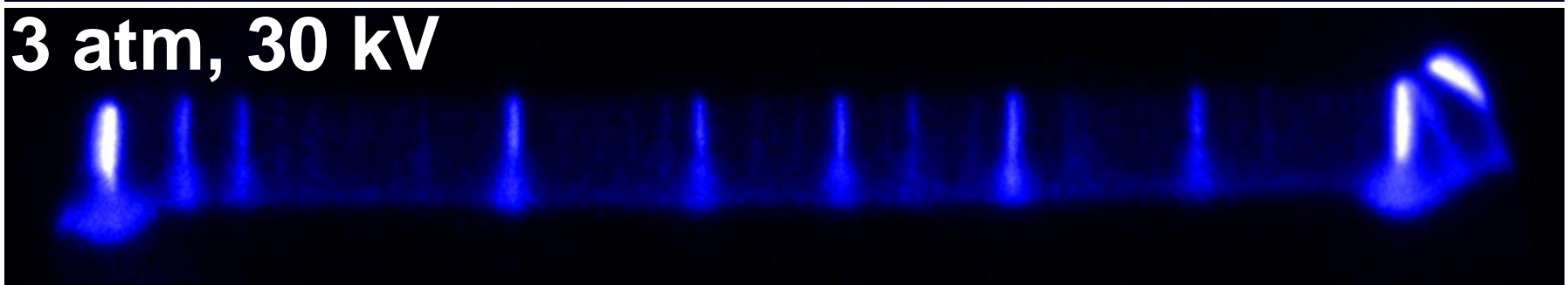
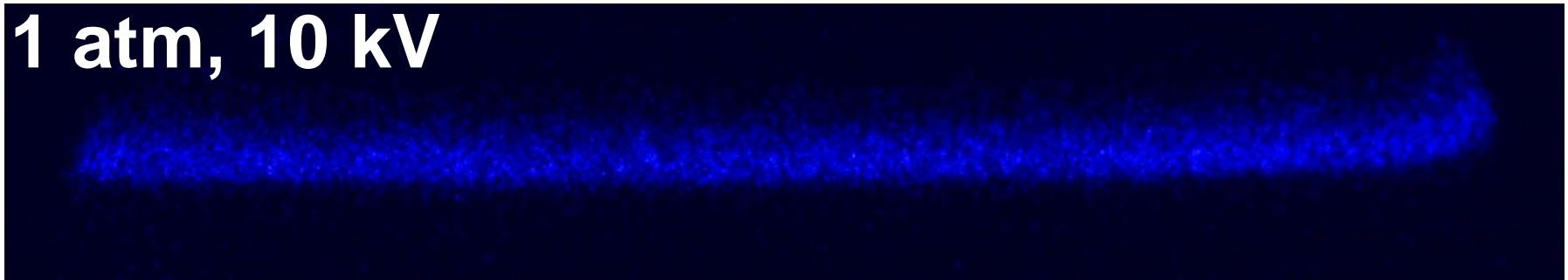
10 kV

30 kV



1 atm, 10 kV

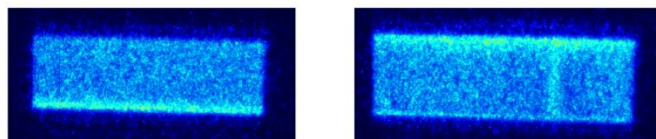
3 atm, 30 kV



DBD Discharges: 20 kV, 10kHz

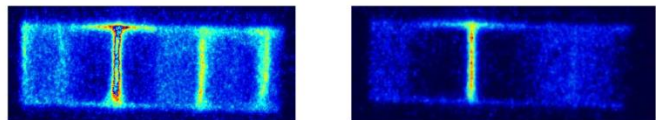
ICCD gate 50 ns

Side view: $T_0=300$ K, $\phi=0.0$, pulse#10



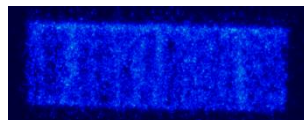
10 Torr

50 Torr

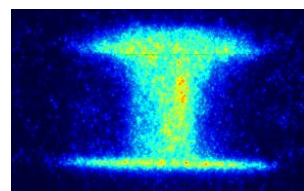


100 Torr

200 Torr



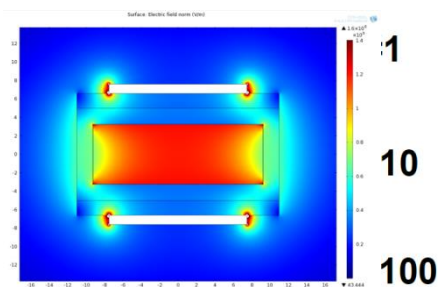
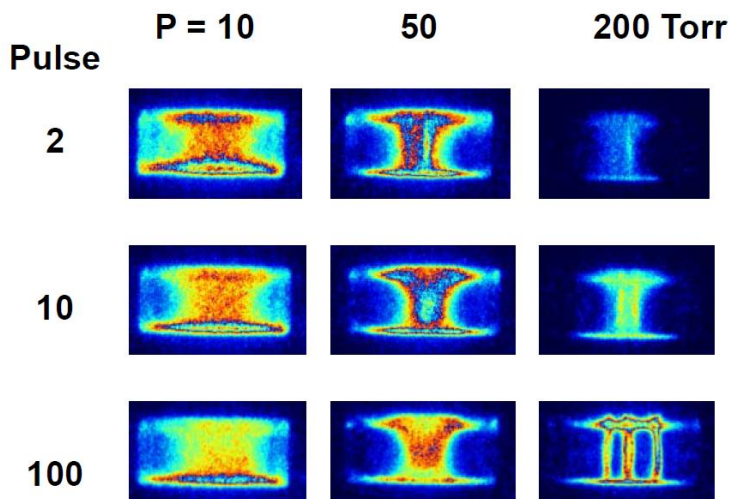
200 Torr



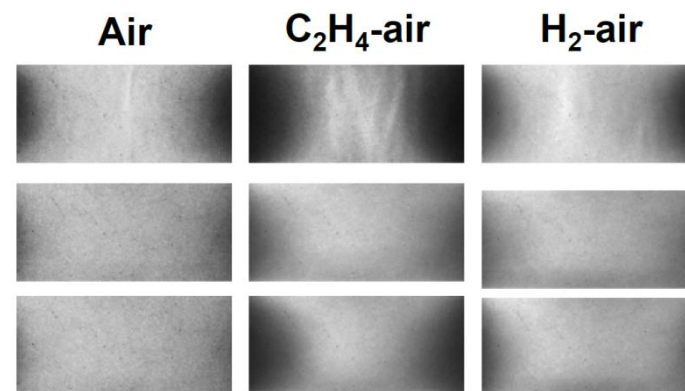
Side view: $T_0=500$ K, $\phi=0.3$

Pressure (torr)	H ₂ -air	C ₂ H ₄ -air
100		
300		
500		

Front view: $T_0=300$ K, $\phi=0.0$



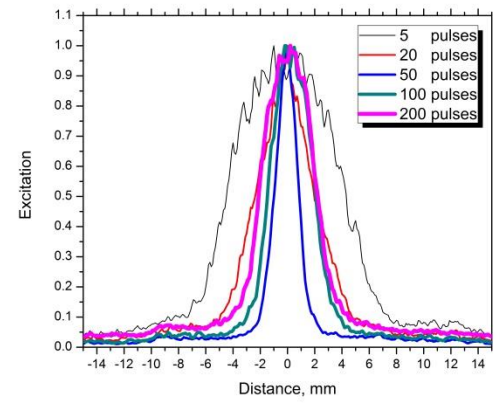
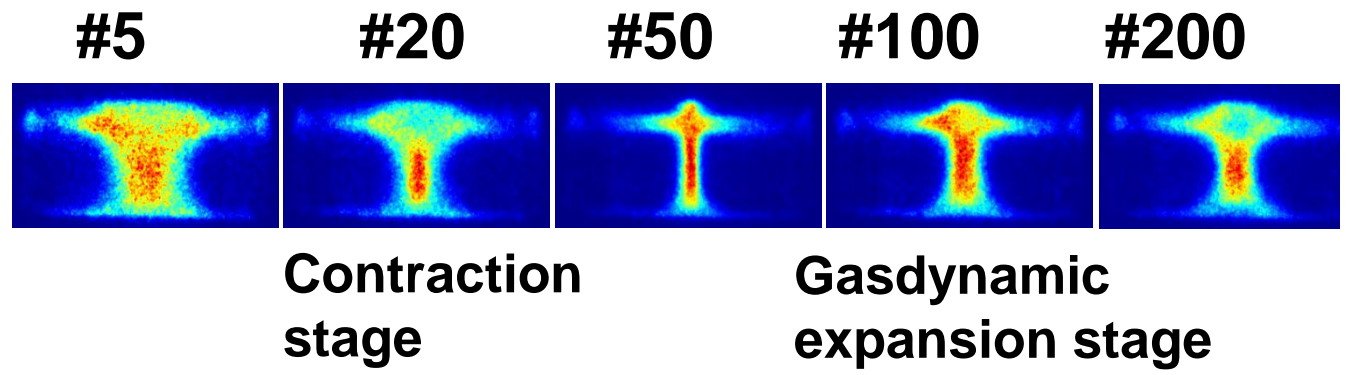
Front view: $T_0=500$ K, $\phi=0.3$



DBD Discharges: 20 kV, 10kHz

ICCD gate 50 ns. P = 20 Torr

Air

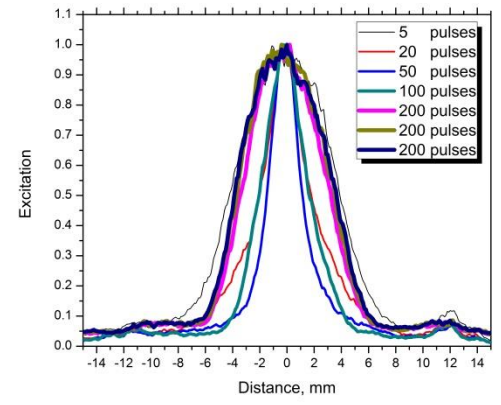
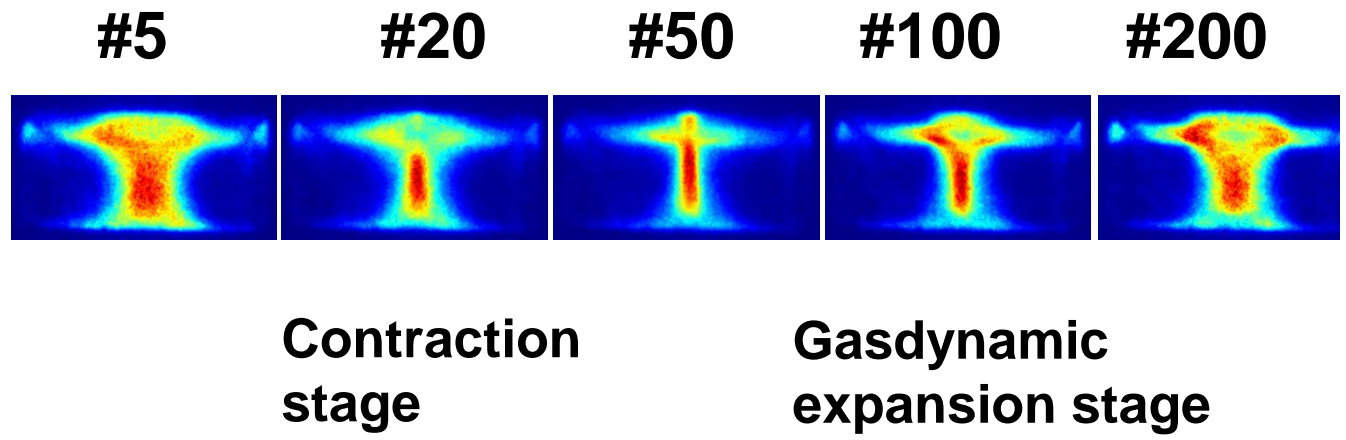


Energy distribution profiles. Dynamic discharge contraction and gasdynamic expansion stages are clearly seen.

$$\tau_{inst} \sim \tau_T / \gamma_i \sim 0.1 \tau_T \quad \gamma_i = (d \ln(v_i)) / (d \ln(E/N)) \quad \tau_{inst} \sim 10^{-4} - 10^{-2} \text{ s}$$

$$\tau_T \sim \gamma / (\gamma - 1) p [\text{Pa}] / \langle W \rangle \quad \text{- typical heating time}$$

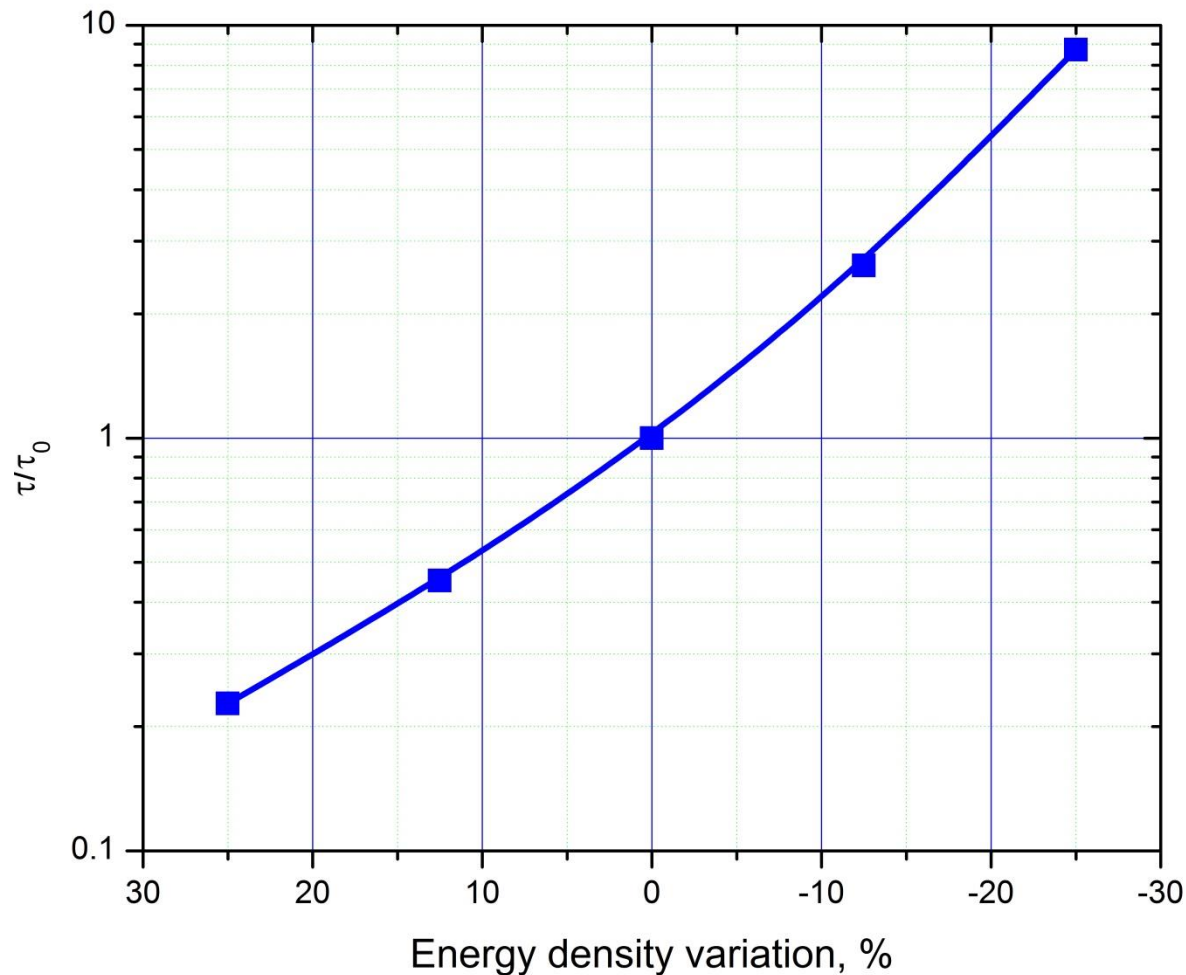
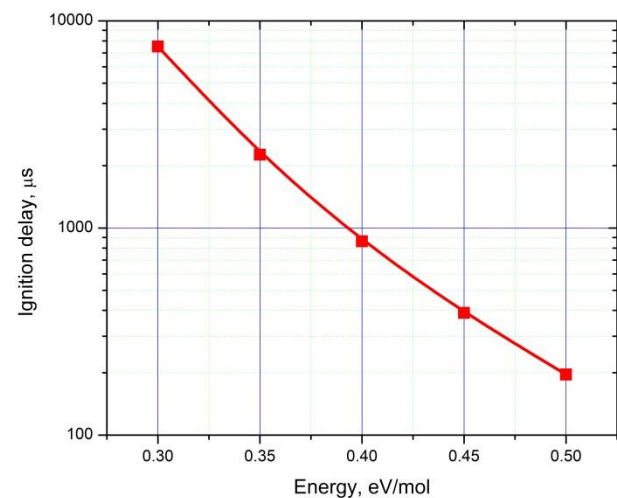
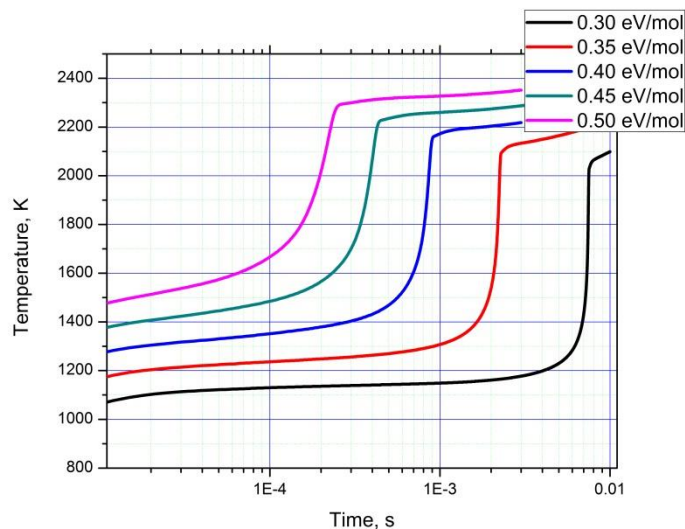
Nitrogen



Kinetic Analysis.

Konnov's Chemical Mechanism, $T_0 = 500\text{K}$, $P = 50\text{ Torr}$

$\text{C}_2\text{H}_6\text{-Air}$. $E/n = 300\text{ Td}$, Different discharge energy.



Even 25% inhomogeneity will lead to order of magnitude difference in ignition delay – and completely compromise the kinetic analysis.

Kinetic Error Analysis

Air

C₂H₄-Air

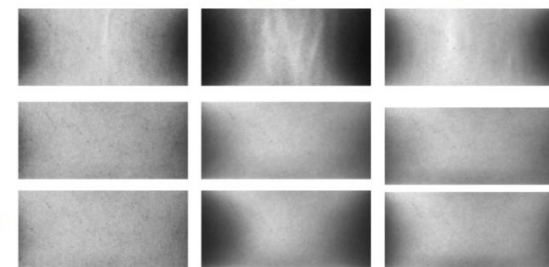
H₂-Air

Air

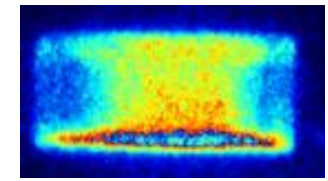
C₂H₄-air

H₂-air

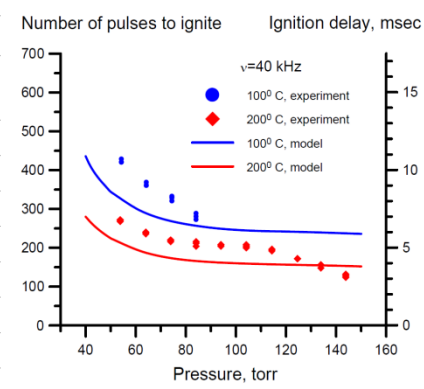
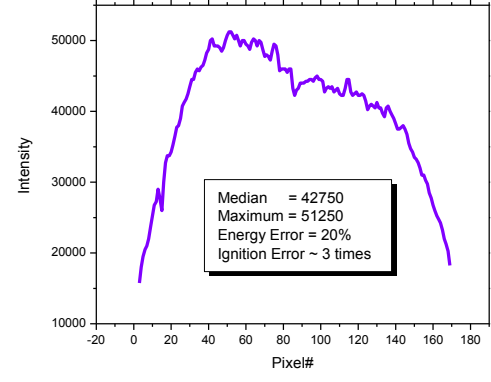
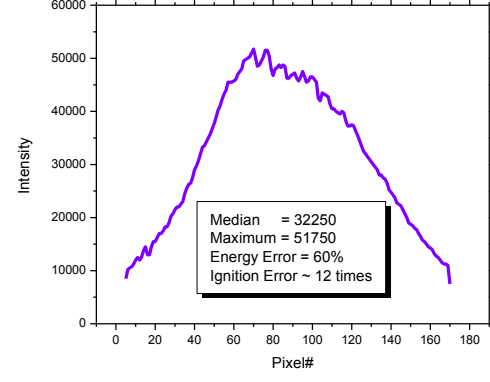
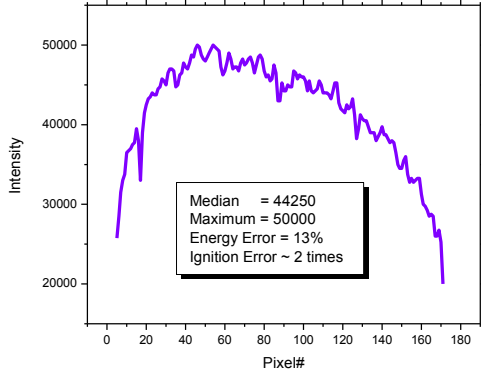
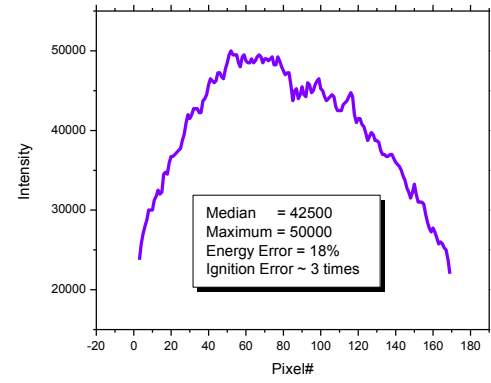
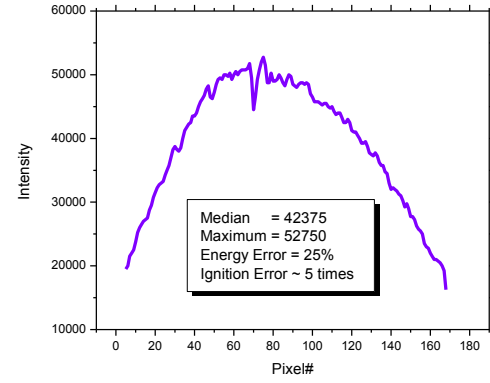
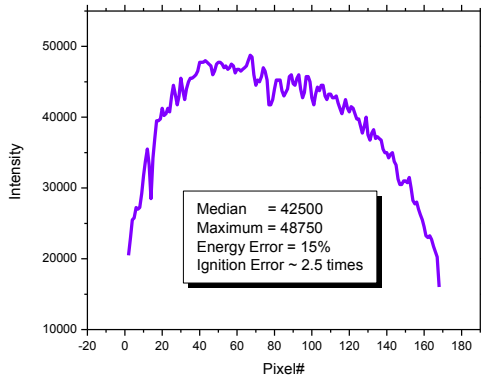
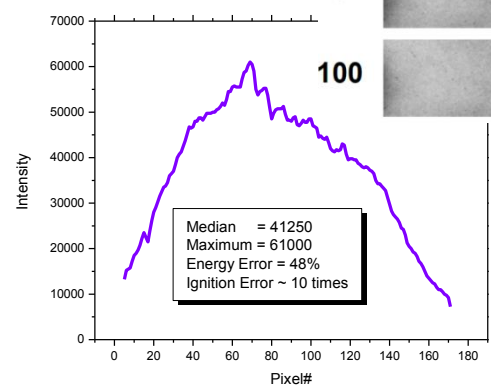
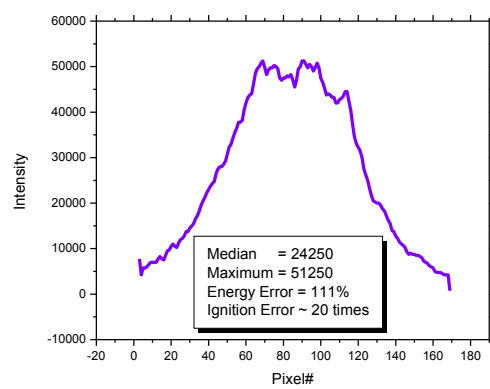
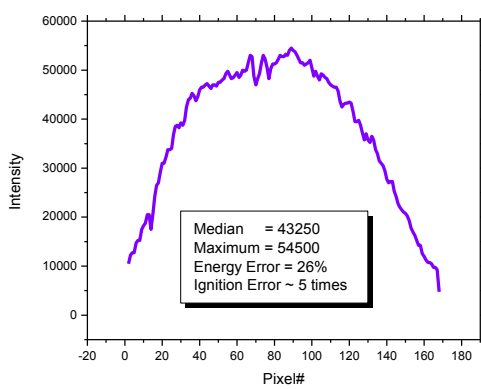
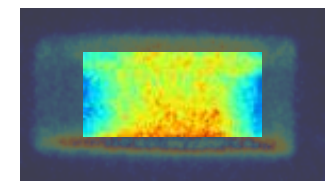
1
10
100



Inhomogeneous

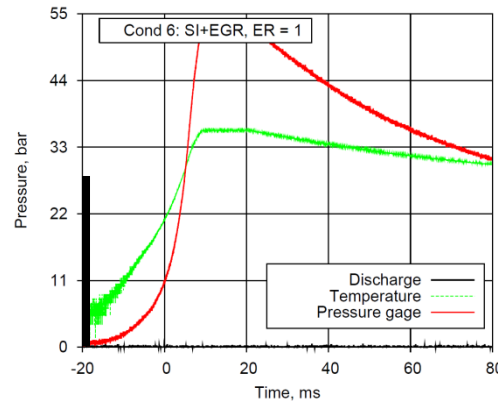
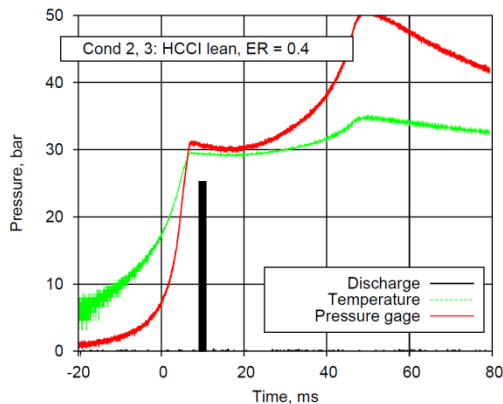
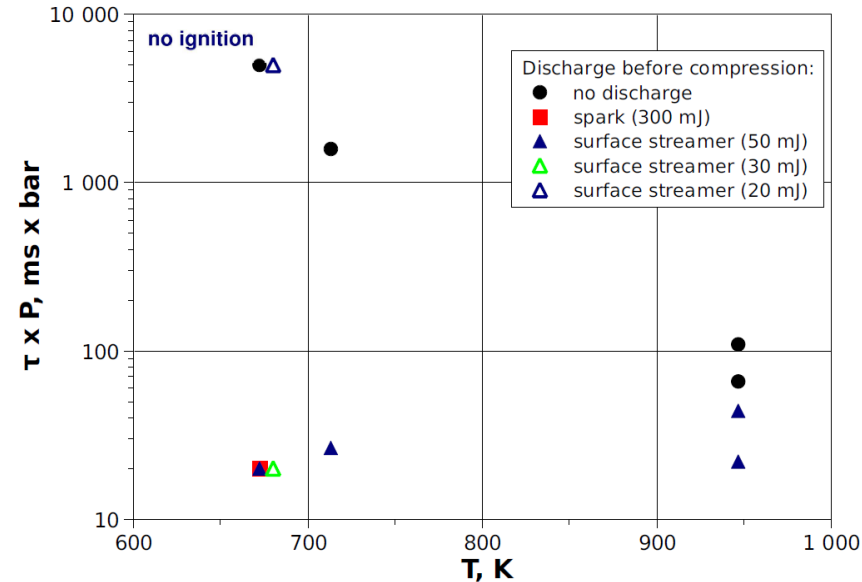
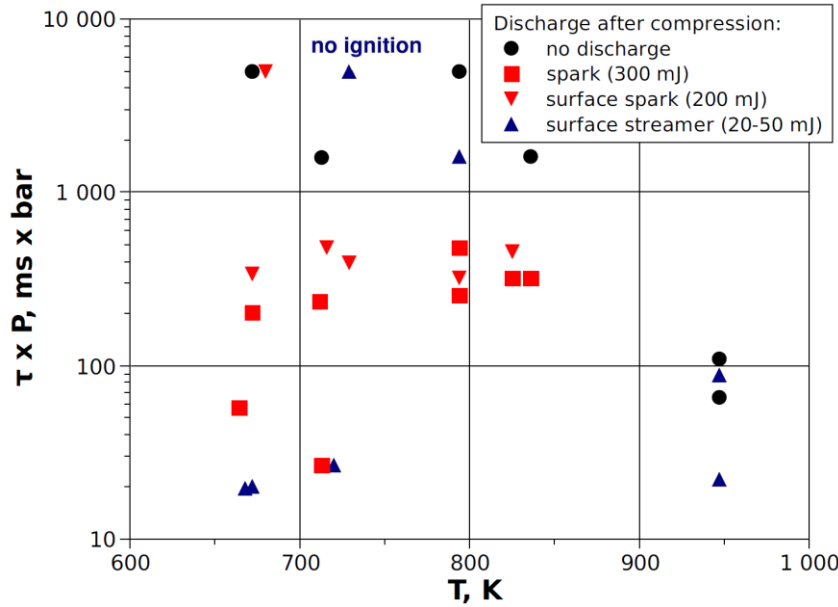
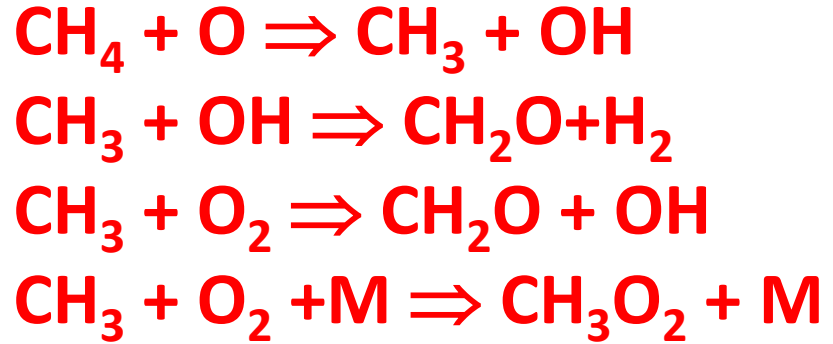


Homogeneous?



Ignition of a stoichiometric hydrogen-air mixture modeling. OSU, 2009

Plasma-Assisted Ignition at High Pressures

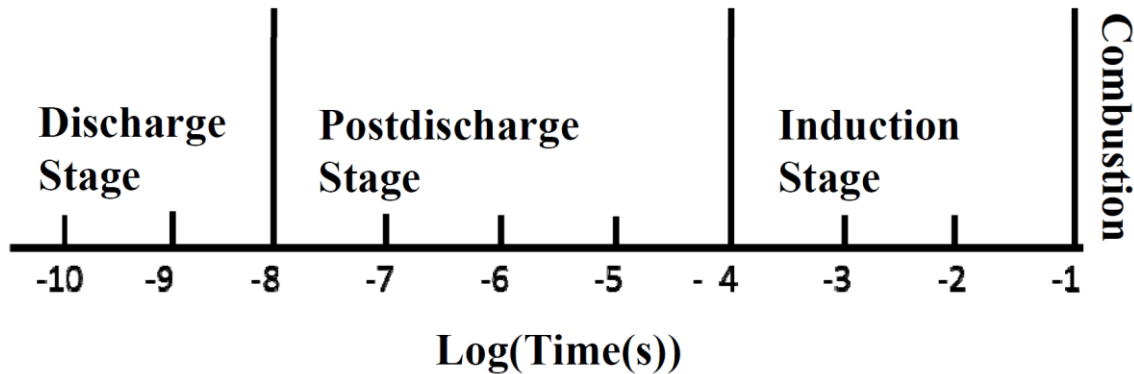


Ignition delay time for modified mixtures, $f=1.0$, EGR=30%. Discharge 20ms before compression stroke

T2 = 794 K, P2 = 32 bar

T2 = 672 K, P2 = 20 bar.

Kinetics of Ignition Development



Stage 1. Discharge in Methane-Air mixture at temperature ~ 330 K, 1 atm. Production of metastable components.

Stage 2. Fast adiabatic compression to a temperature of 800-950 K. Metastable components decomposition and ignition development.

CH ₂ O	CO	CH ₃ OH	CH ₃ O ₂ H	H ₂ O ₂	Delay Time	Sensitivity
0	0	0	0	0	1.00	
540ppm	170 ppm	260 ppm	21 ppm	49 ppm	0.33	
540 ppm					0.51	910
	170 ppm				1.00	0
		260 ppm			0.89	423
			21 ppm		0.60	19,050
				49 ppm	0.47	10,820

alkylperoxy radicals!

Non-diffusive hybrid scheme for simulation of filamentary discharges

FLUID MODEL

The balance equation within hydrodynamic (drift-diffusion) approximation for required species and Poisson's equation for electric potential:

$$\begin{aligned}\frac{\partial n_s}{\partial t} + \text{div } \vec{j}_s &= Q_s & \Delta\varphi &= -\frac{1}{\epsilon\epsilon_0} \sum q_s n_s \\ \vec{j}_s &= \vec{W}_s n_s - D_s \nabla n_s & \vec{E} &= -\nabla\varphi \\ \vec{W}_s &= \mu_s \vec{E}\end{aligned}$$

Secondary processes of electron production: photoionization in $\text{N}_2\text{-O}_2$, ion-electron emission, photoemission.

HYBRID MODEL

Non-fluid regions: $N_s = n_s \times \Delta V \leq 1$

1. DISCRETE FLUXES

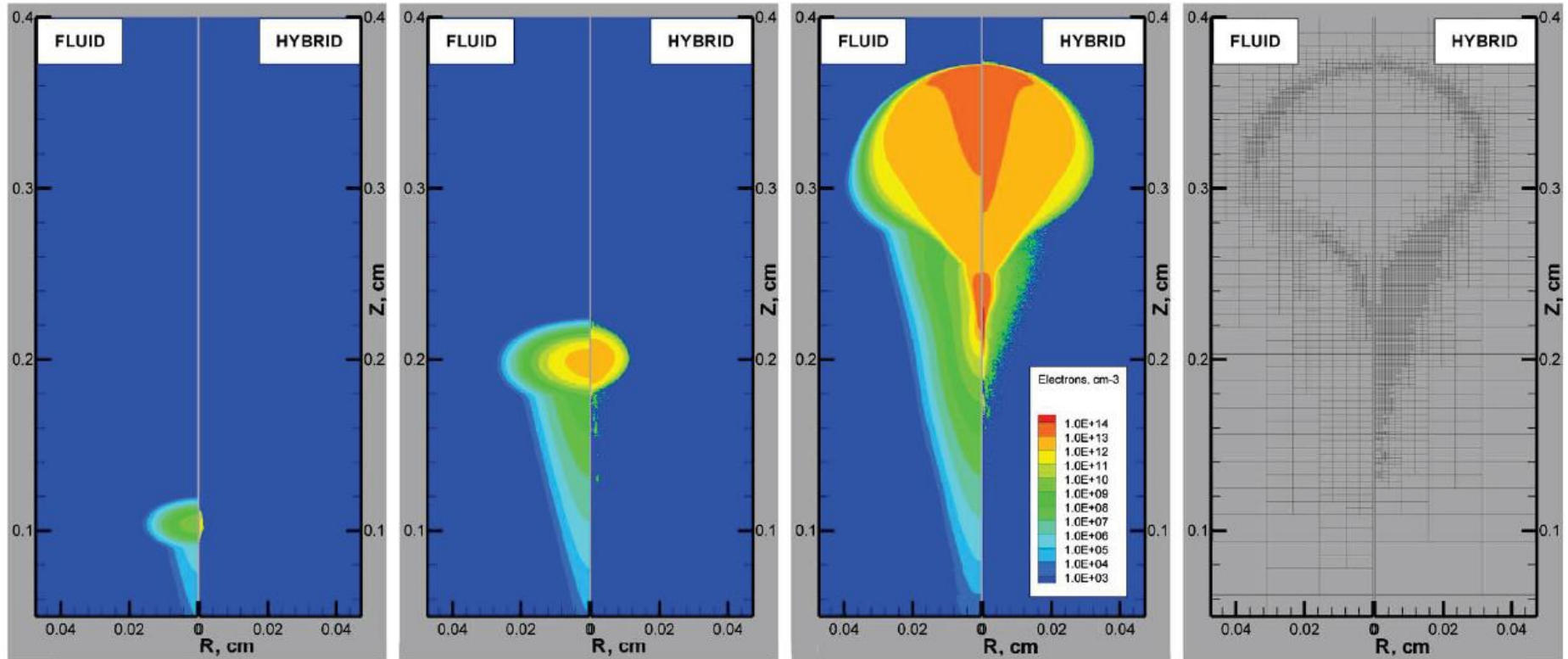
Original flux j_x and number ΔN of transported over interface A_x species:

$$j_x = W_x n_s - D \frac{\partial n_s}{\partial x} \Rightarrow \Delta N = |j_x| A_x \Delta t$$

$$\Delta N = \Delta N^{int} + \Delta N^{rem},$$

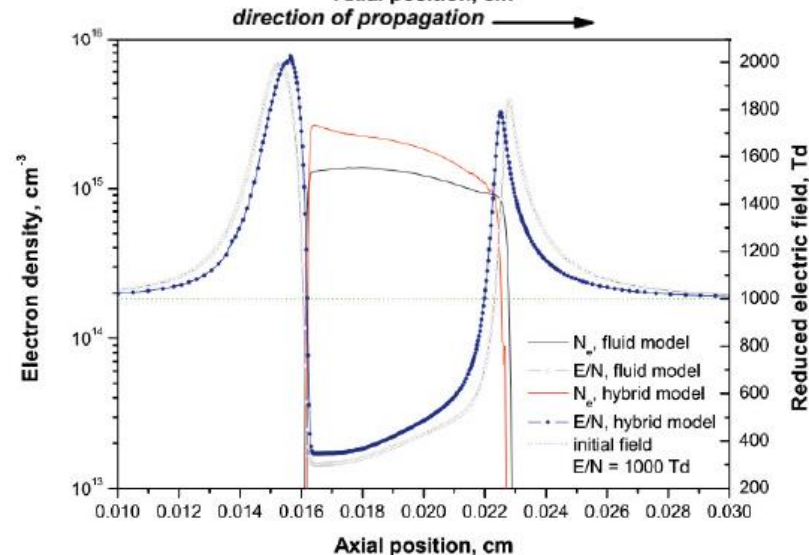
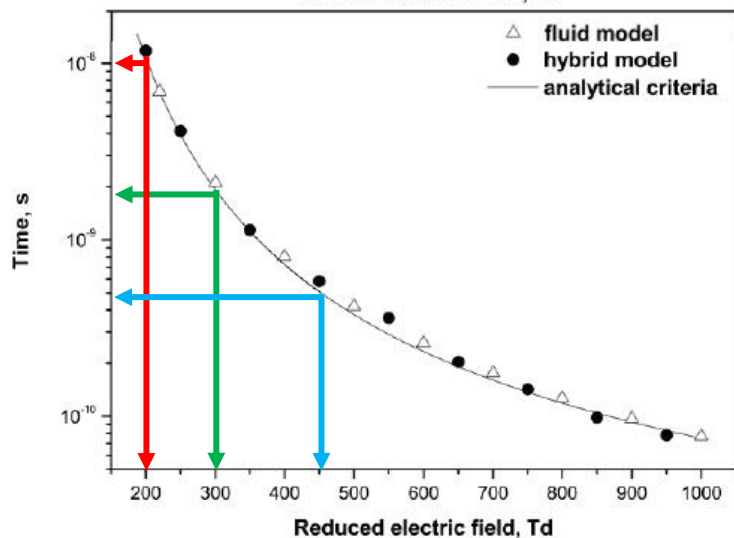
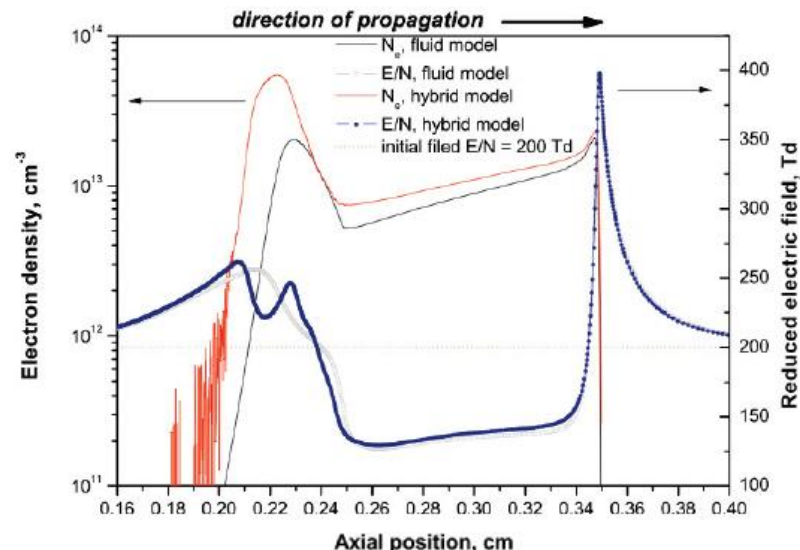
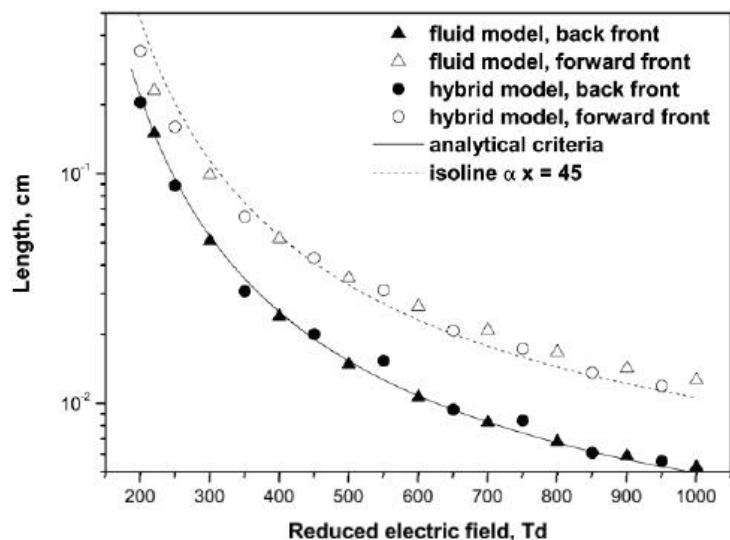
$$\text{where } \Delta N^{int} \in \mathbb{Z} \text{ and } \Delta N^{rem} < 1$$

AVALANCHE TO STREAMER TRANSITION IN UNIFORM ELECTRIC FIELD (air, 1 bar, 300 K, 1 cm, various voltages)



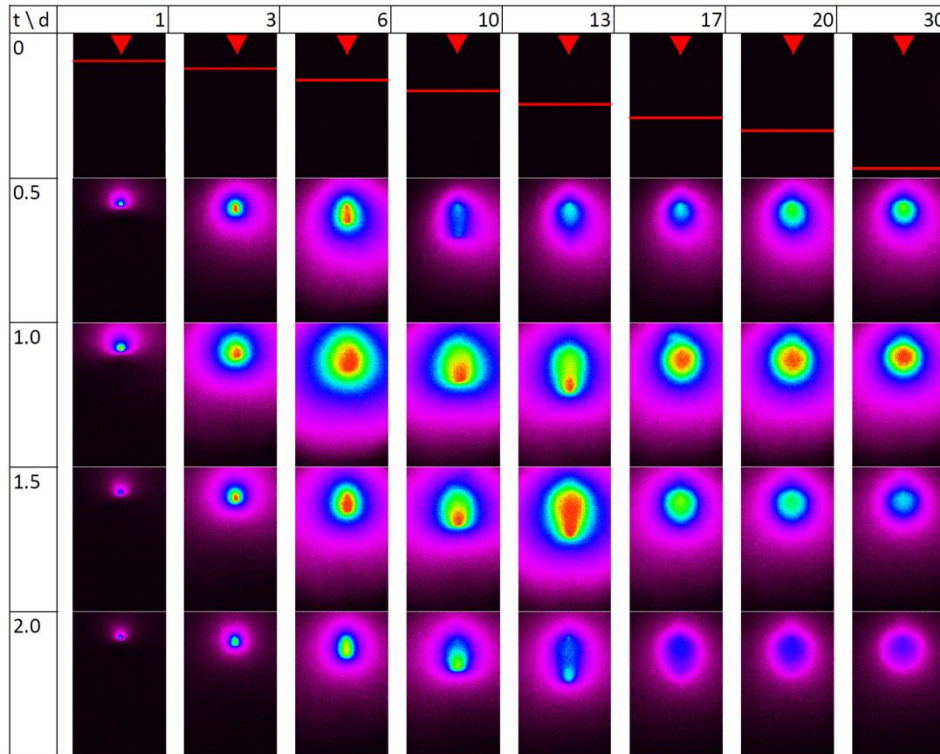
AVALANCHE TO STREAMER TRANSITION IN UNIFORM ELECTRIC FIELD

(air, 1 bar, 300 K, 1 cm, various E/n)



PS High-Pressure Discharge

Air, 1 atm



$$E/n = 200 \text{ Td}: \tau_{\max}(\rho_0) \sim 10 \text{ ns}$$

$$E/n = 300 \text{ Td}: \tau_{\max}(\rho_0) \sim 2 \text{ ns}$$

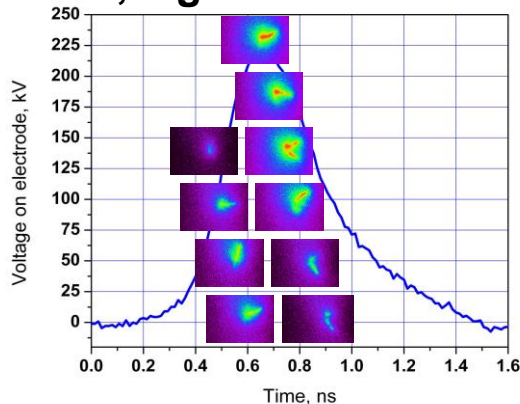
$$\rho_1 \sim 21\rho_0 (P_1 = 70 \text{ atm})$$

$$\tau_{\max}(200 \text{ Td}) \sim 500 \text{ ps}$$

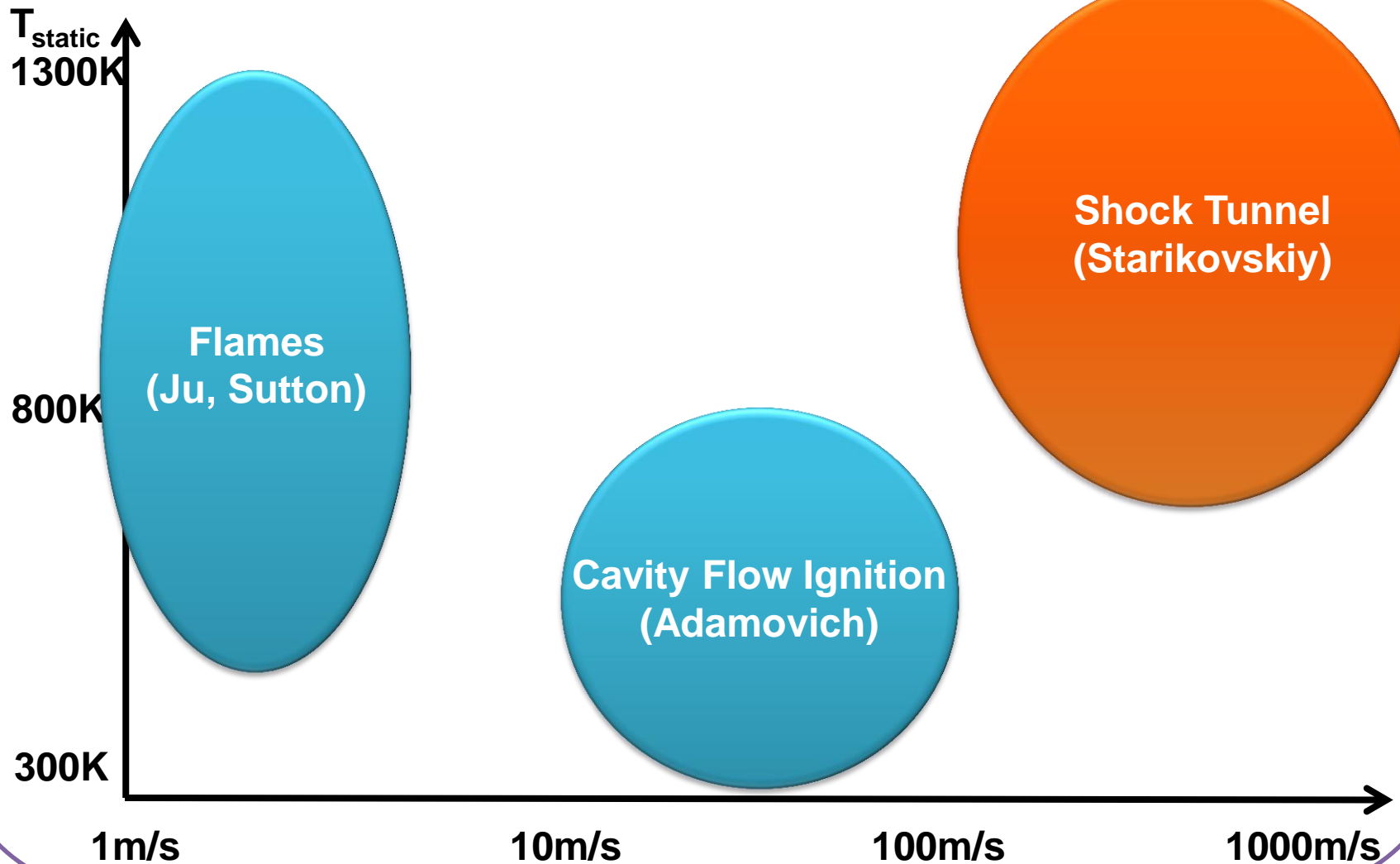
$$\tau_{\max}(300 \text{ Td}) \sim 100 \text{ ps}$$

FPG 200-01PB pulse generator
Voltage up to 200 kV
Pulse duration 350 ps
Rise time 120-140 ps
Voltage rise rate $2 \times 10^{15} \text{ V/s}$

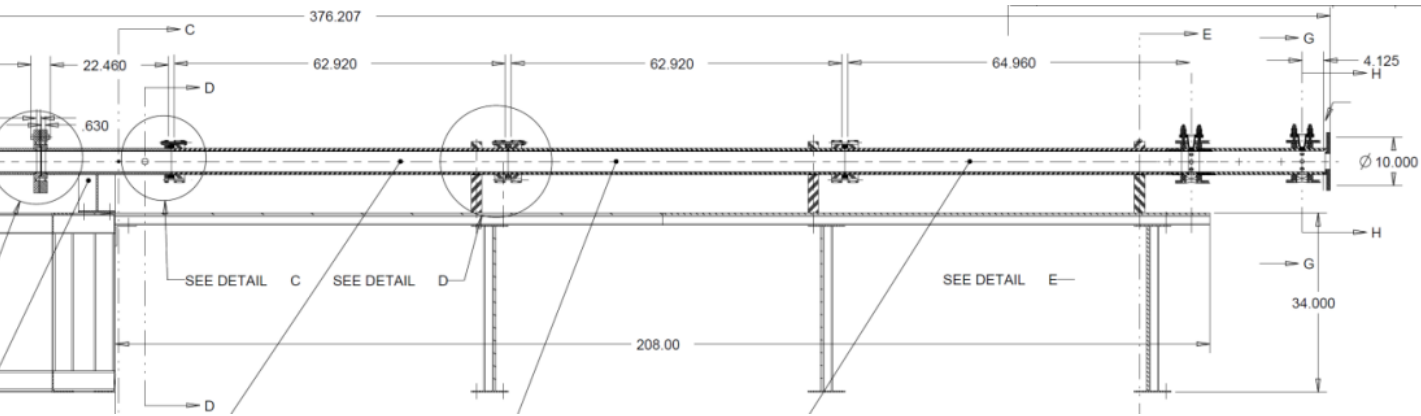
Water, 1 g/cm³



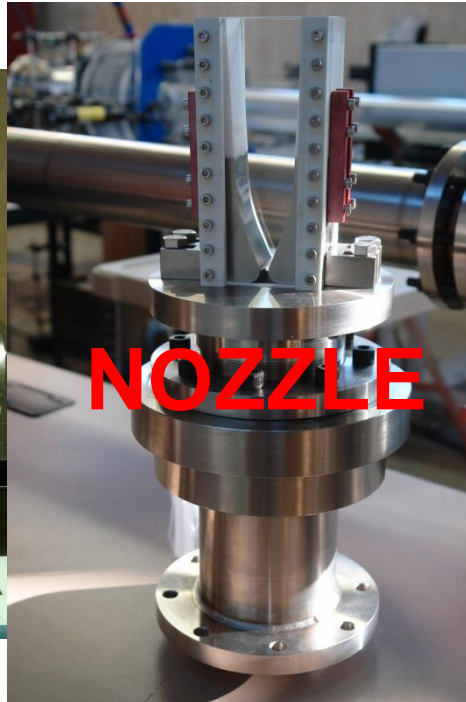
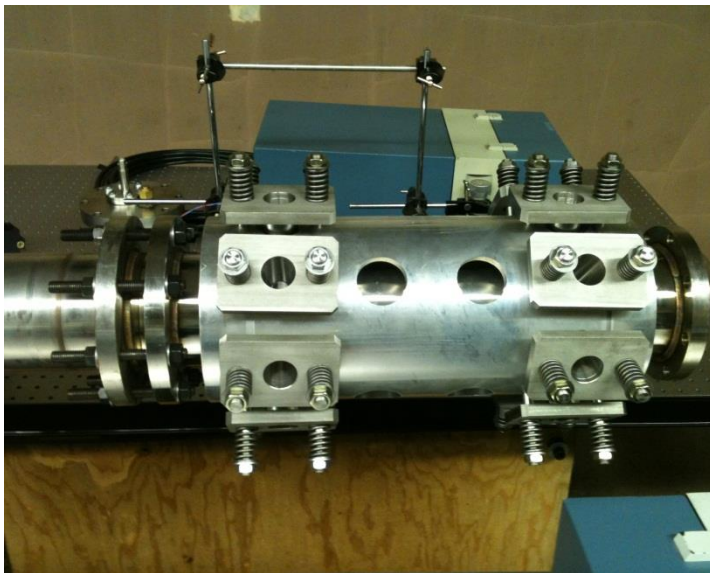
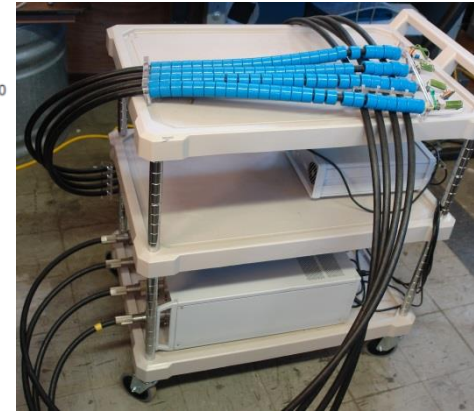
MURI Deliverables: Diffusion, Mixing, Transport and High-Speed Combustion



Discharge Formation and Flame Stabilization in High Speed Flow – Plasma Shock Tunnel



Pulser

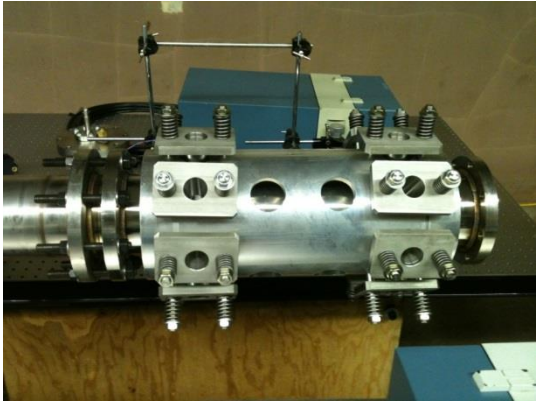


Vacuum Chamber 1x0.5x0.5 m³

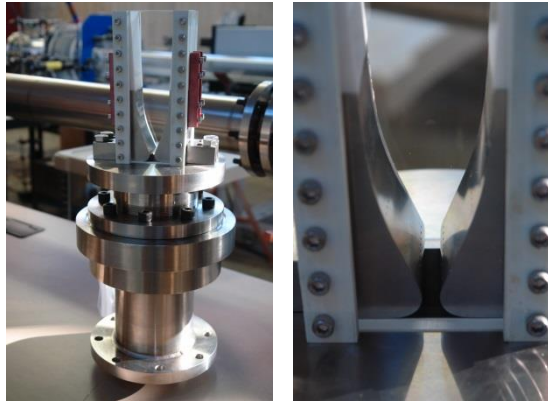
Combustion-Driven Shock Tube

Discharge Formation and Flame Stabilization in High Speed Flow – Plasma Shock Tunnel

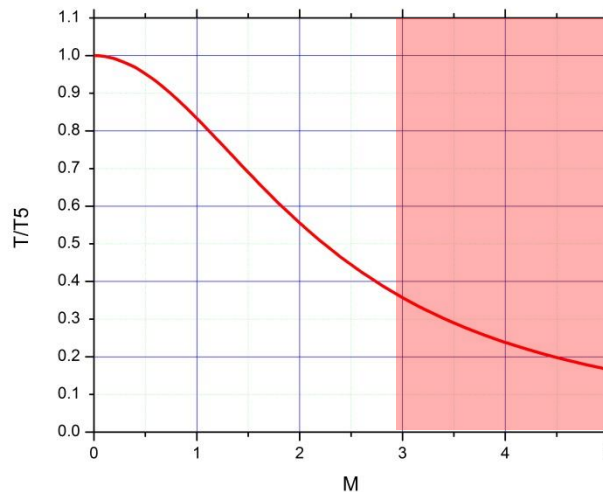
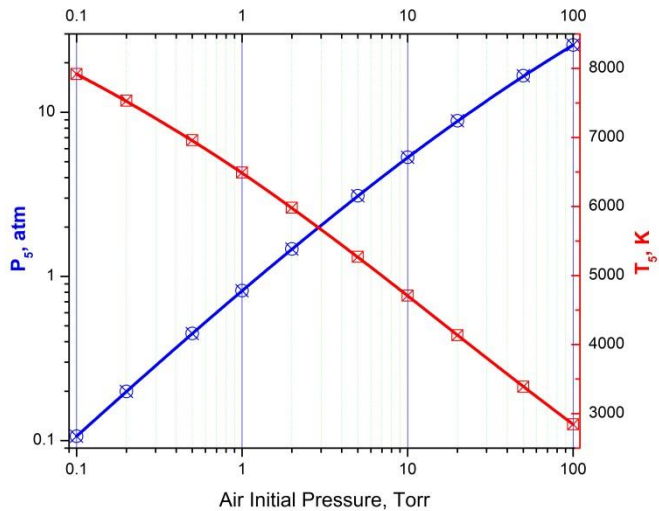
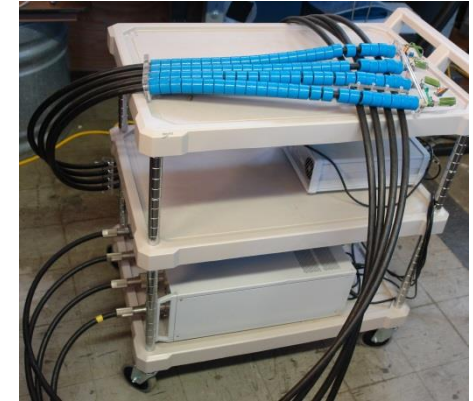
Combustion-Driven Shock Tube



Nozzle

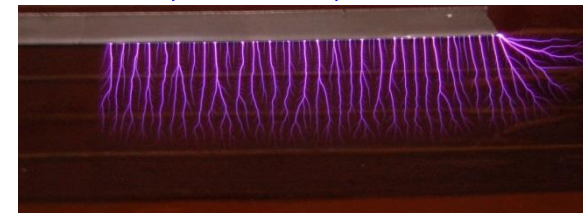


Pulser

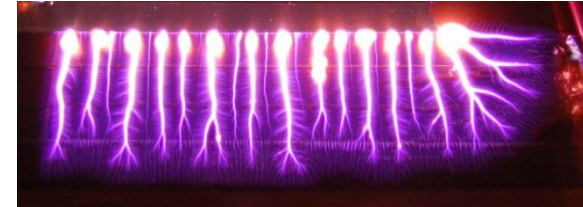


100 kV, 1 MHz
12 ns, 1000 p/b

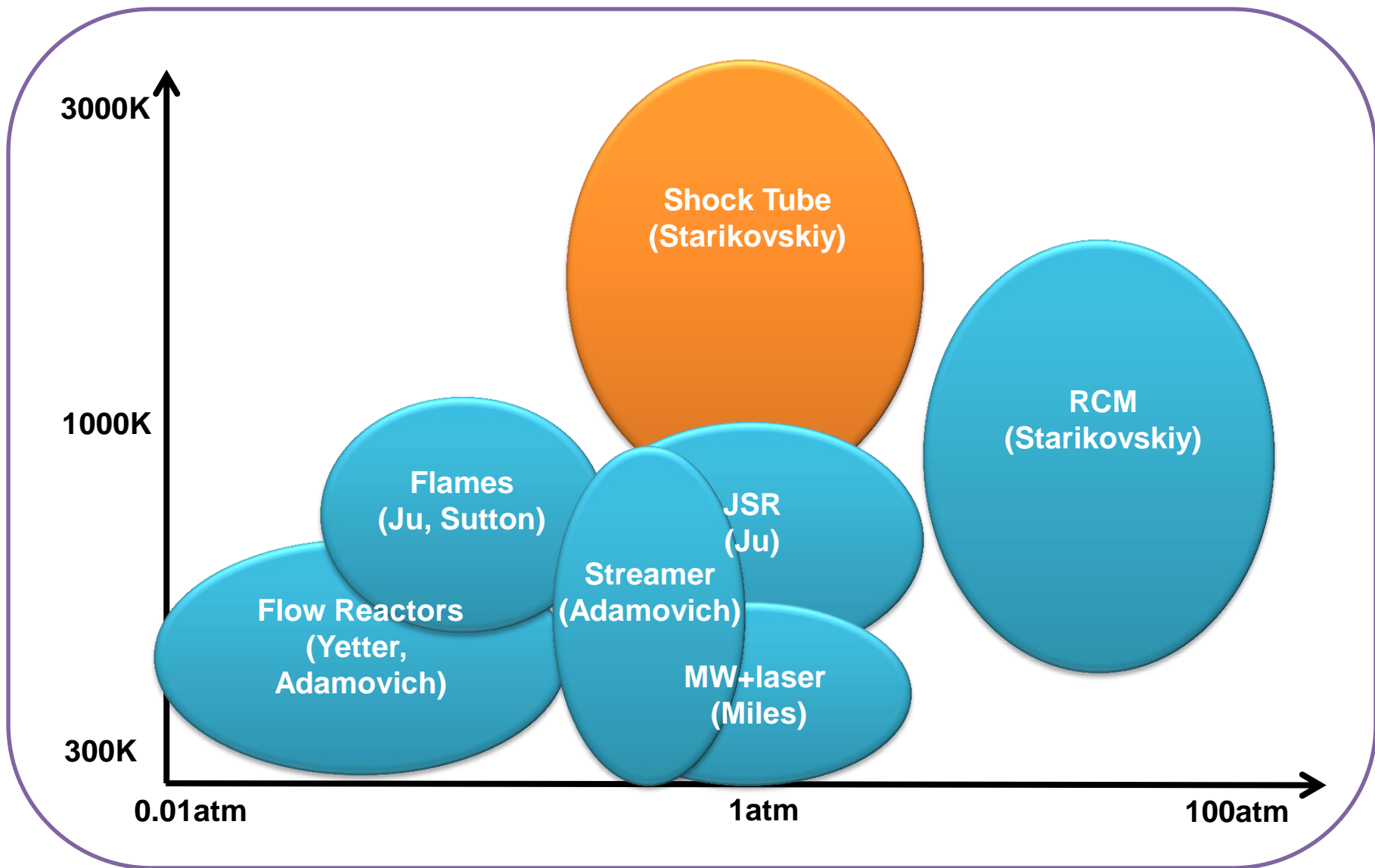
1 MHz, 50 kV, 1 ms



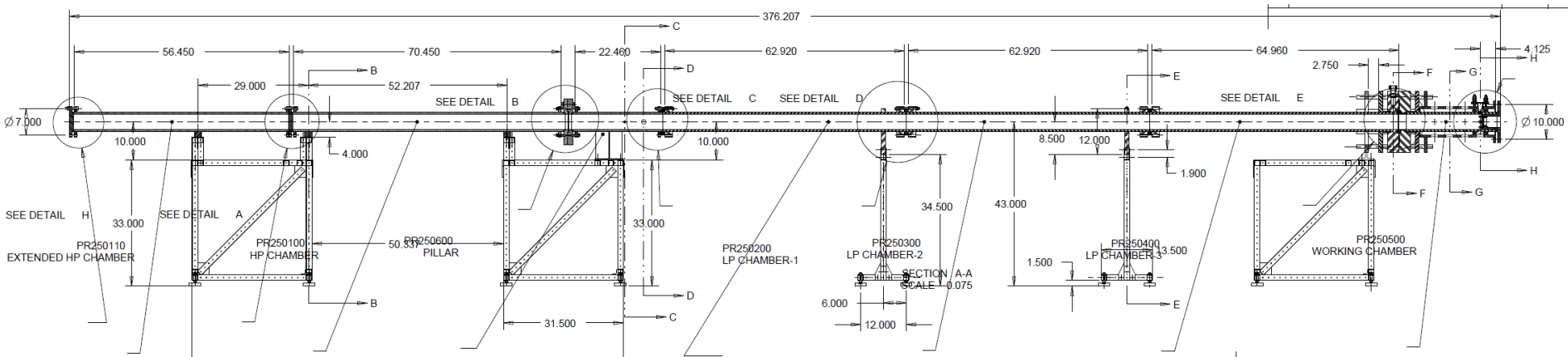
1 MHz, 100 kV, 1 ms



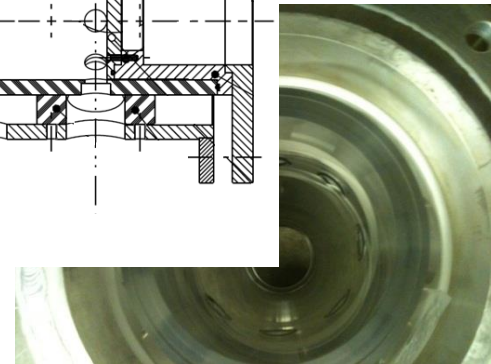
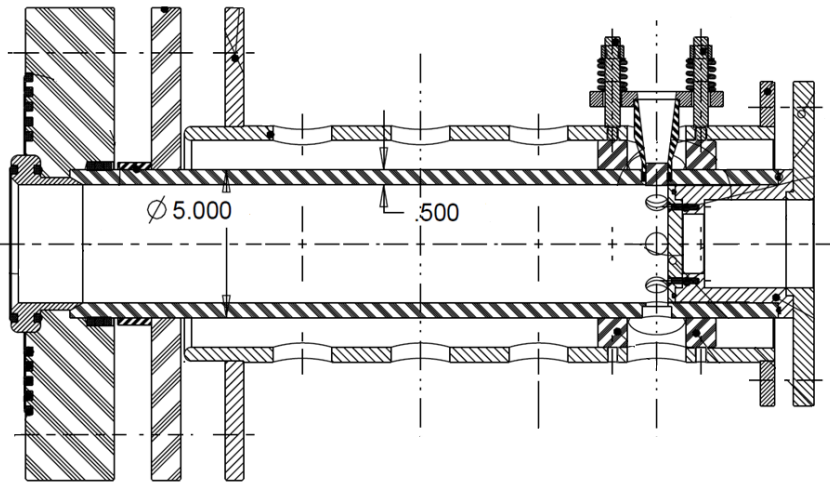
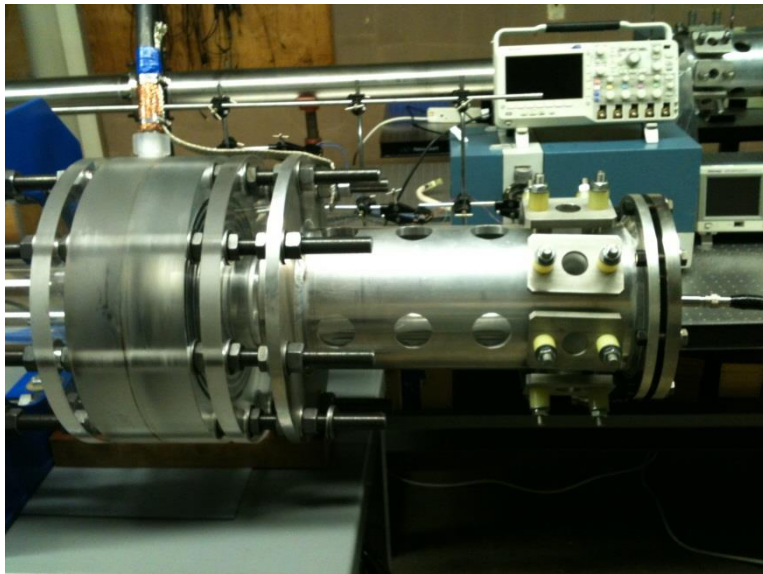
MURI Deliverables: Kinetic Data Generation



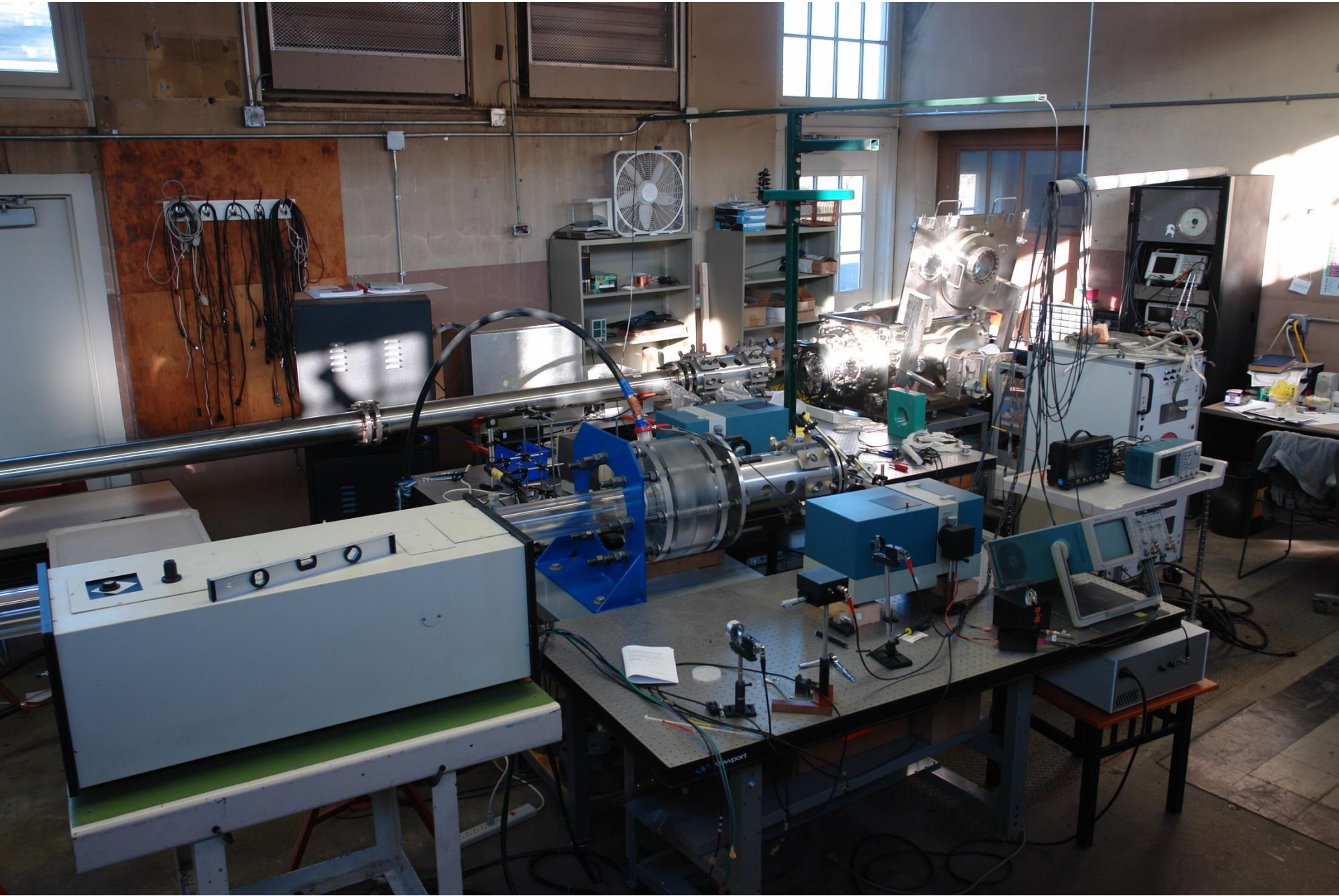
Plasma Shock Tube



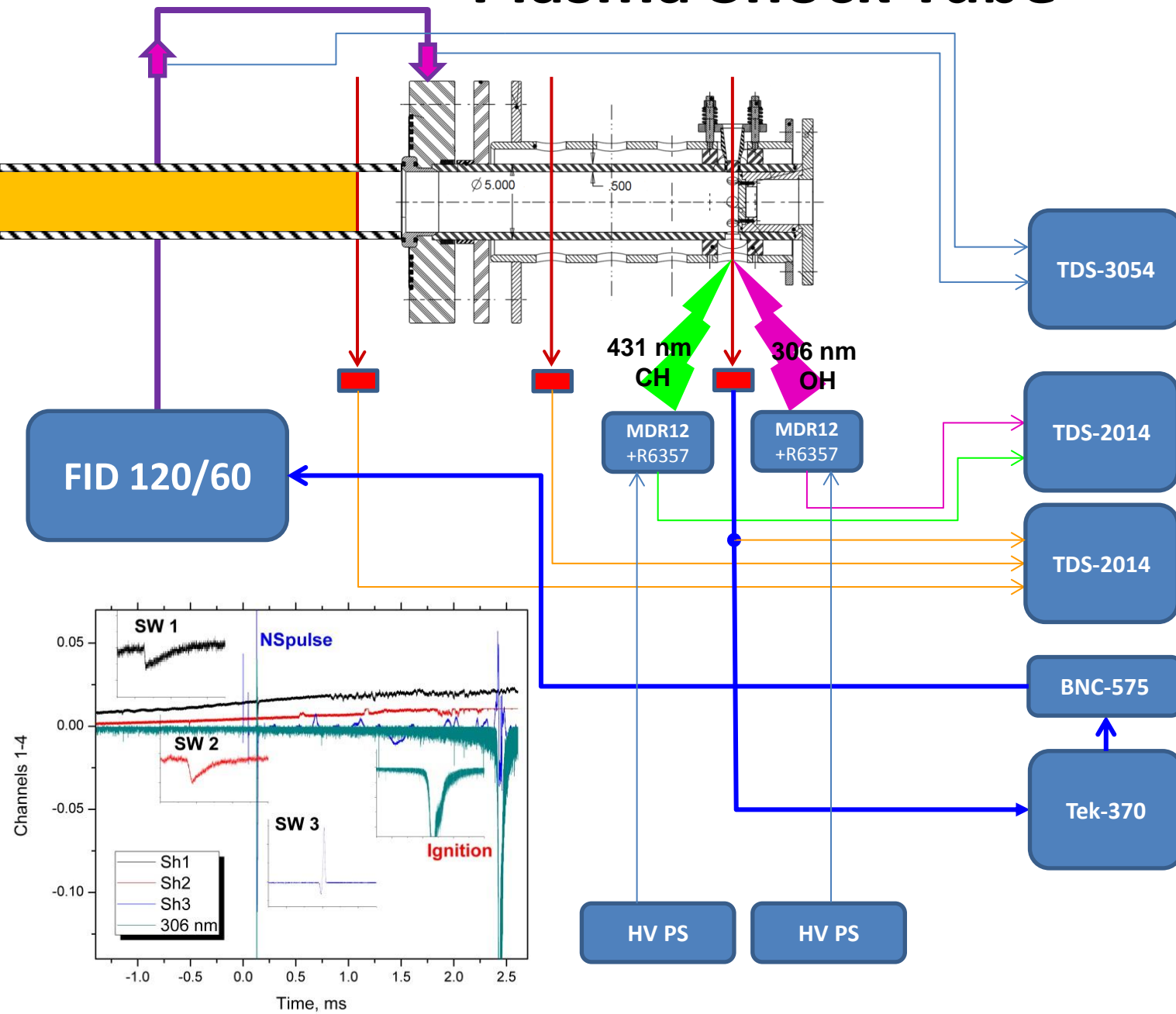
PAC Kinetics at High T, Low P



Plasma Shock Tube

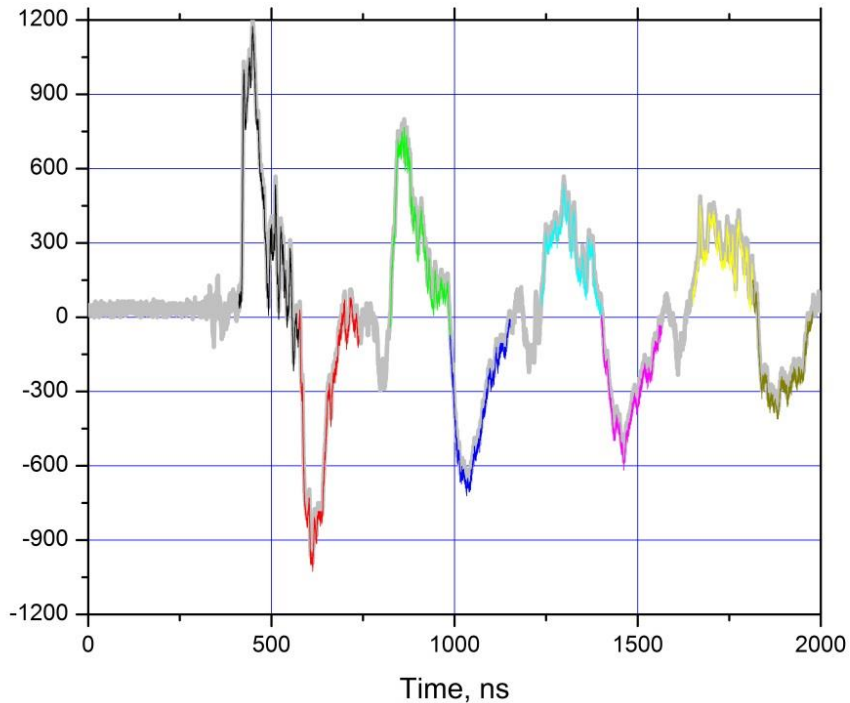


Plasma Shock Tube



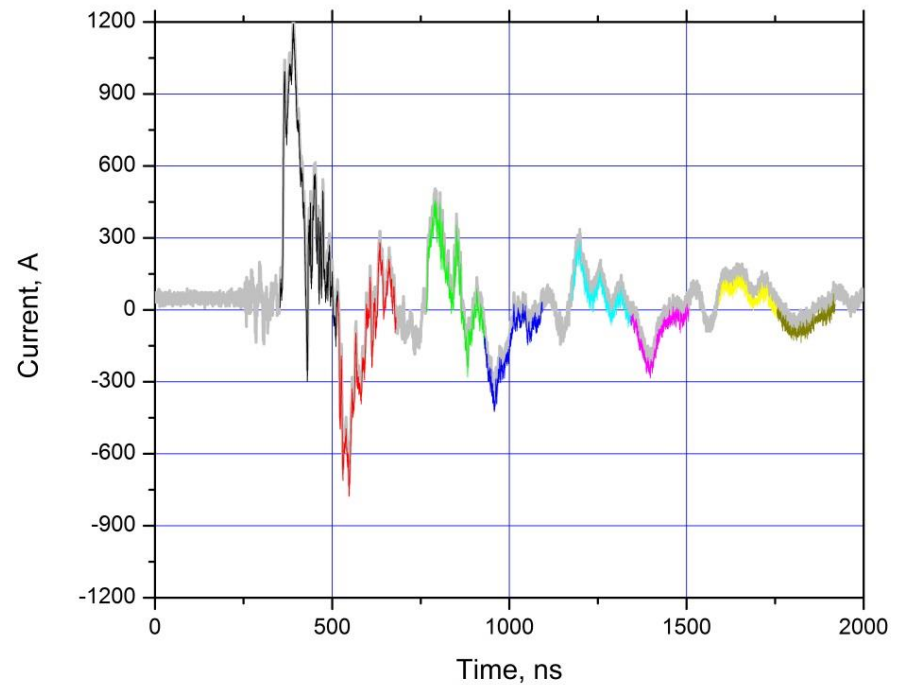
Pulse Current Dynamics – Cable

$C_2H_6:O_2:N_2:Ar = 2:7:28:63$



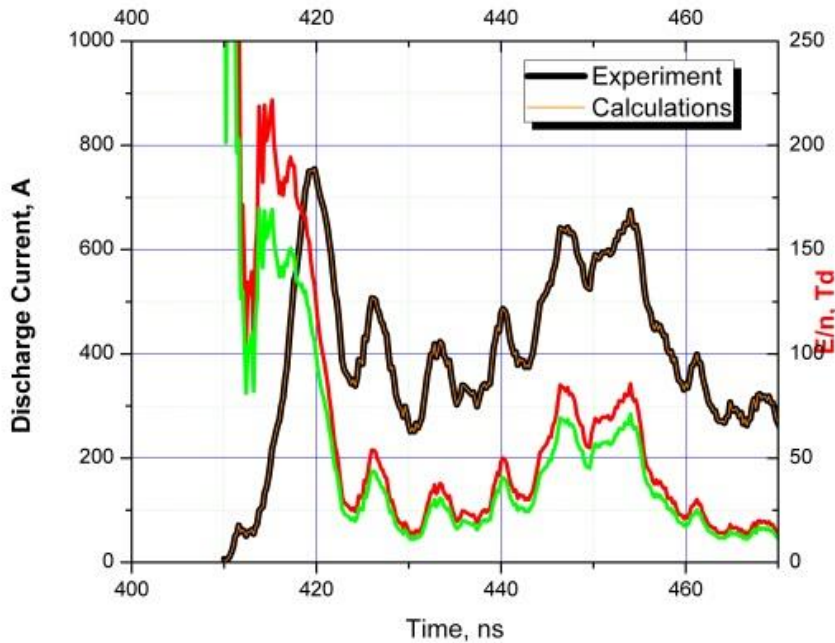
$P_5 = 3.3 \text{ atm}$
 $T_5 = 1360 \text{ K}$
 $\rho_5 = 1.06 \text{ kg/m}^3$

$P_5 = 1.0 \text{ atm}$
 $T_5 = 1610 \text{ K}$
 $\rho_5 = 0.273 \text{ kg/m}^3$

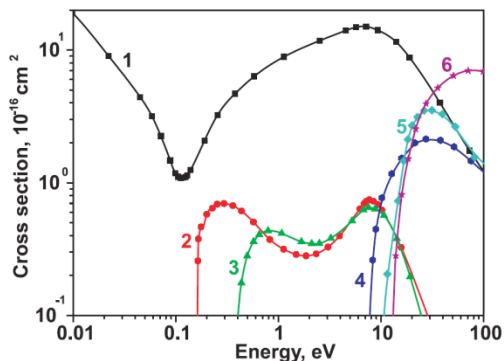


Pulse Current Dynamics – Shock Tube

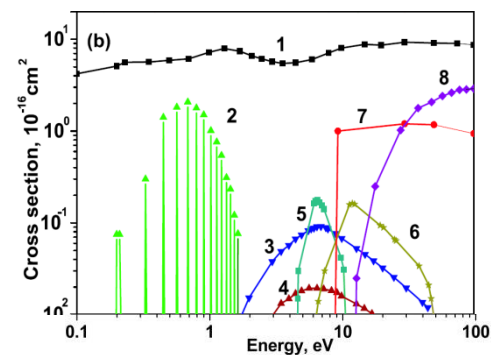
$C_2H_6:O_2:N_2:Ar = 2:7:28:63$



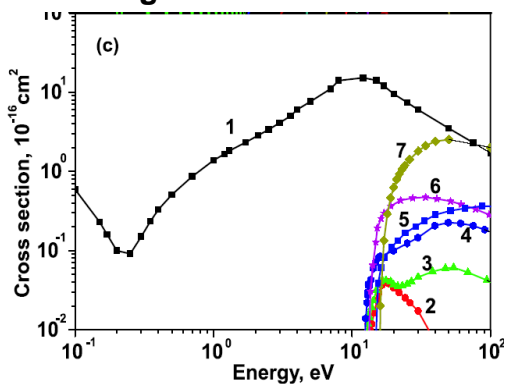
Ethane. Hayashi 1987



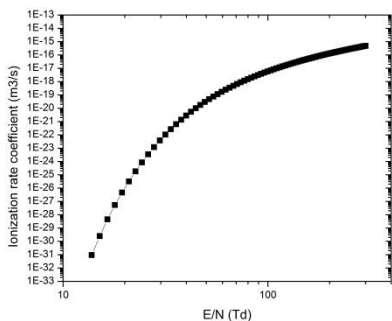
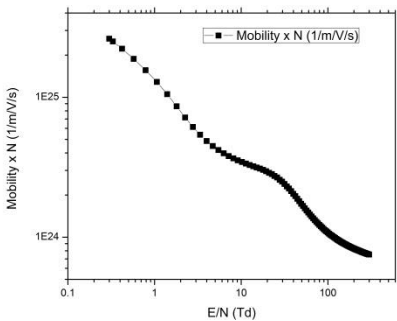
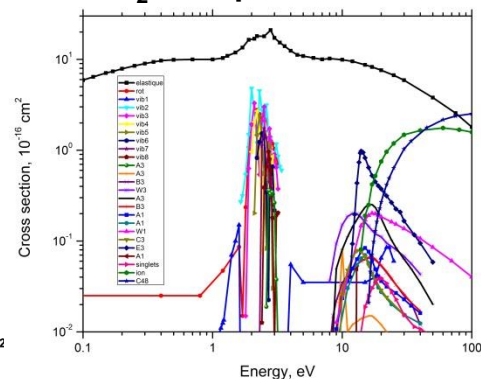
Oxygen. Ionin 2007



Argon. Tachibana 1989



N_2 . Phelps 1994

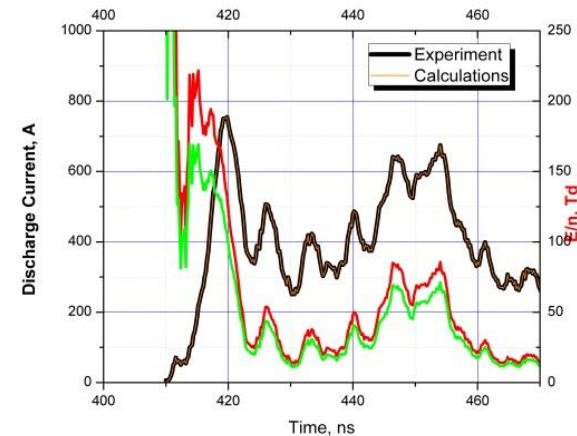
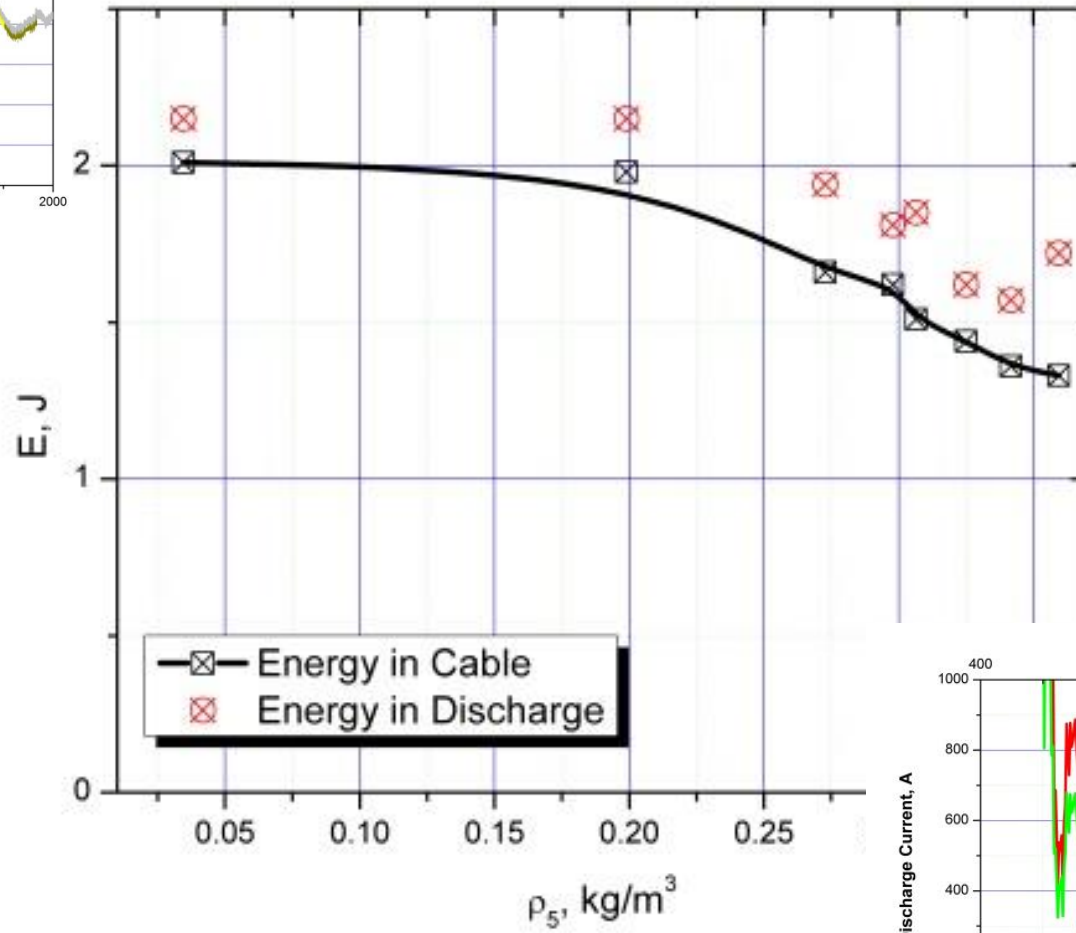
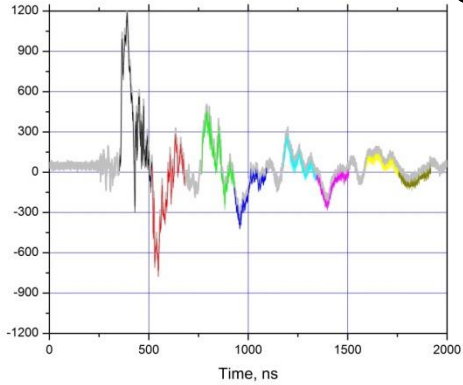


$$\frac{\partial(nf)}{\partial t} + \mathbf{v} \cdot \nabla(nf) + \frac{Ze}{m} \{ \mathbf{E} + \frac{1}{c} [\mathbf{v} \times \mathbf{H}] \} \cdot \nabla_v(nf) = S(nf)$$

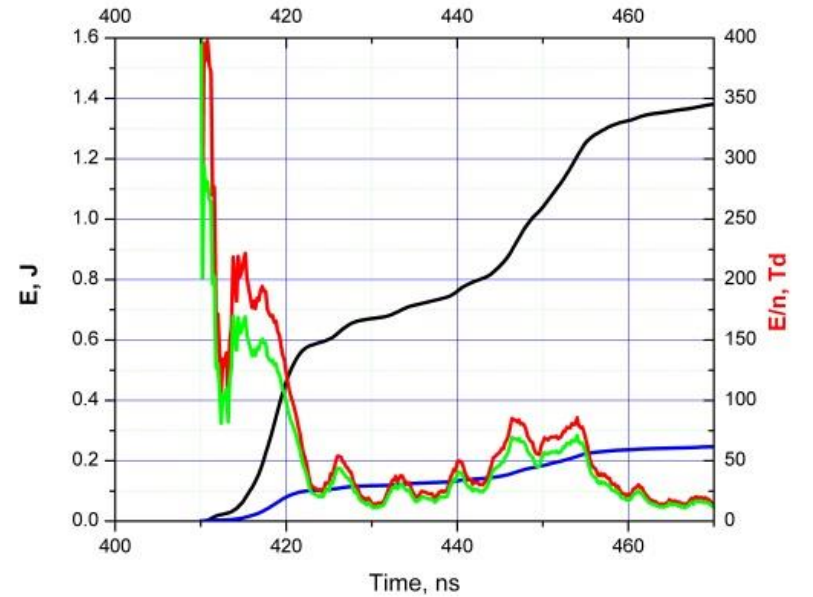
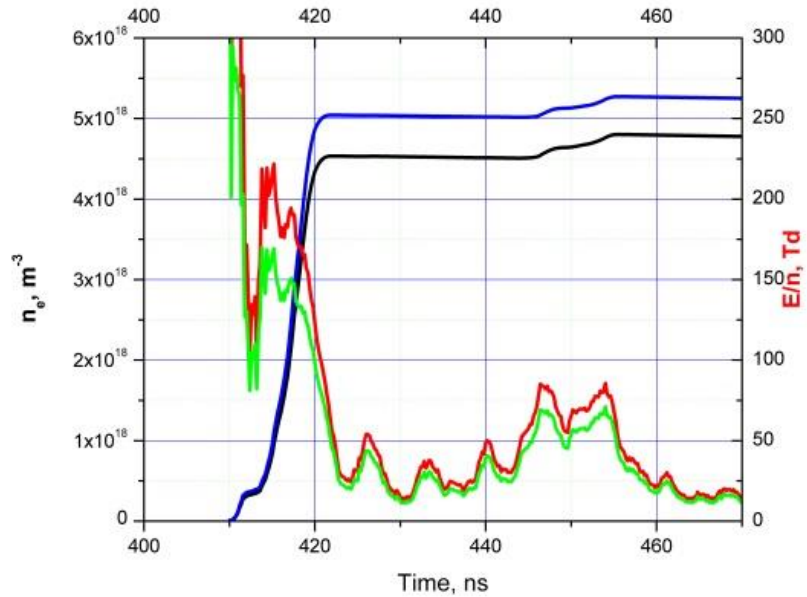
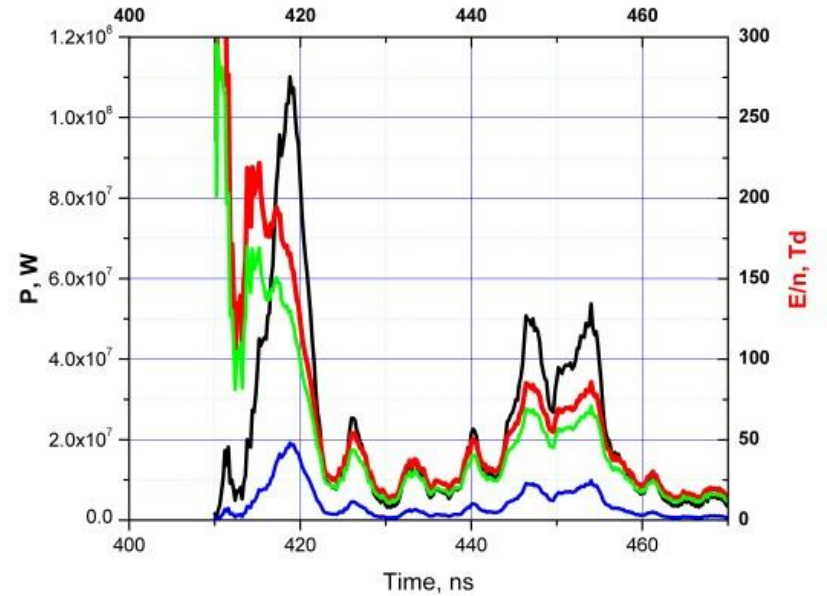
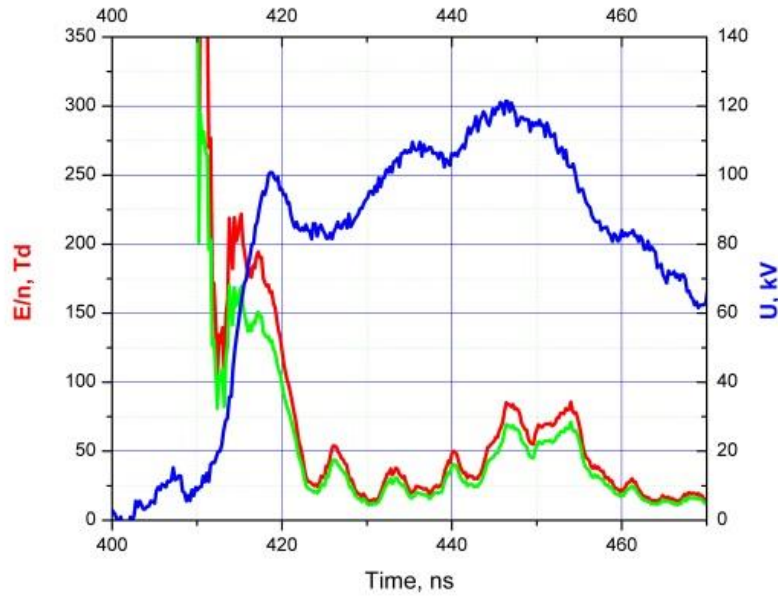
$$f(v, \theta) = \sum_{l=0}^{\infty} f_l(v) P_l(\cos \theta) \approx f_0(v) + f_1(v) \cos \theta \quad v_E/v_m \ll 1$$

Discharge Energy Comparison

$C_2H_6:O_2:N_2:Ar = 2:7:28:63$

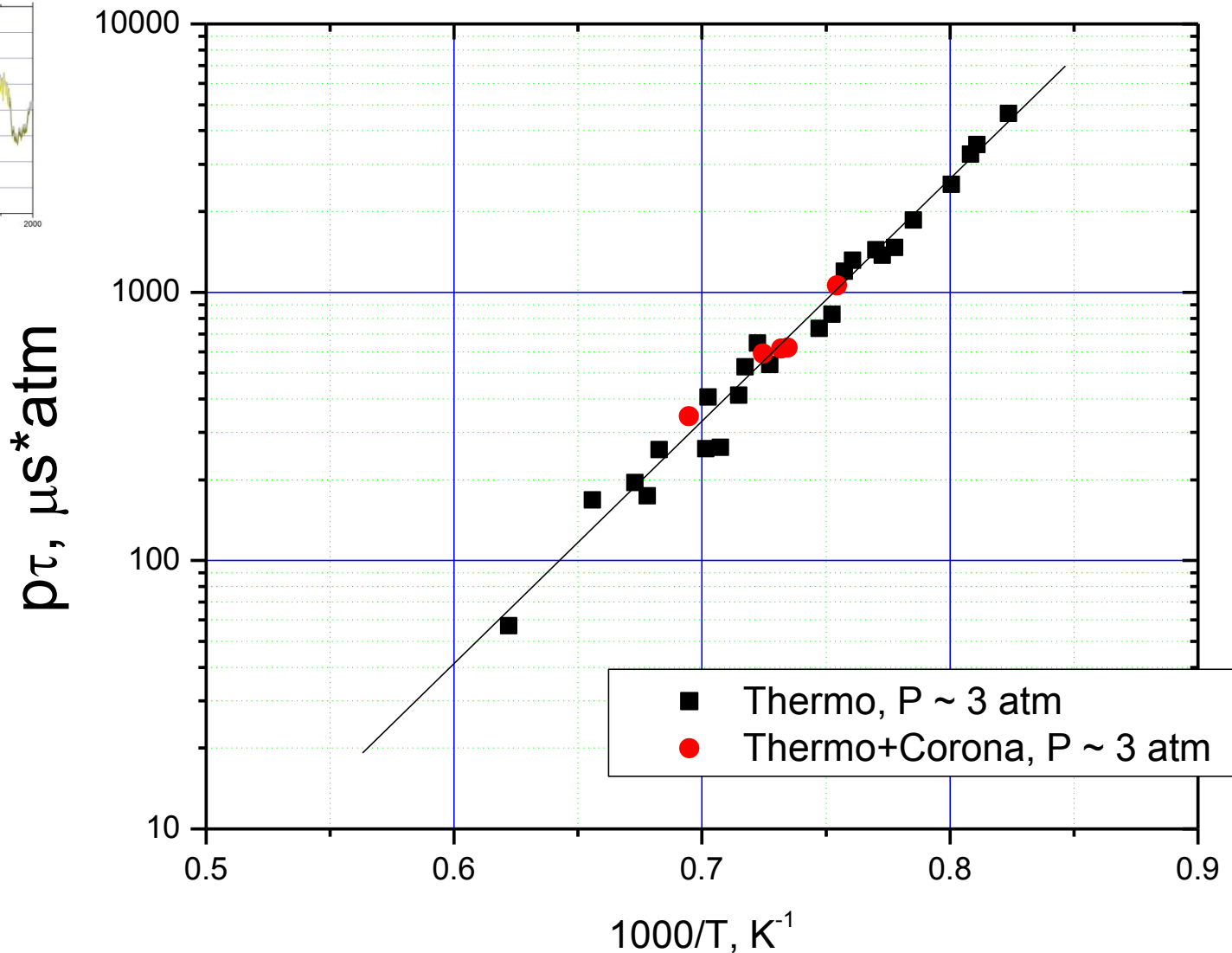
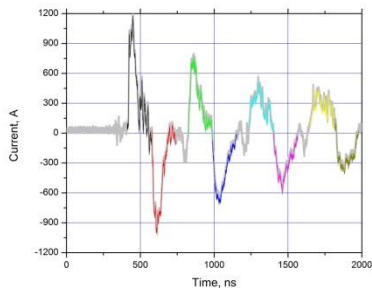


Discharge Dynamics



Ignition Delay Time

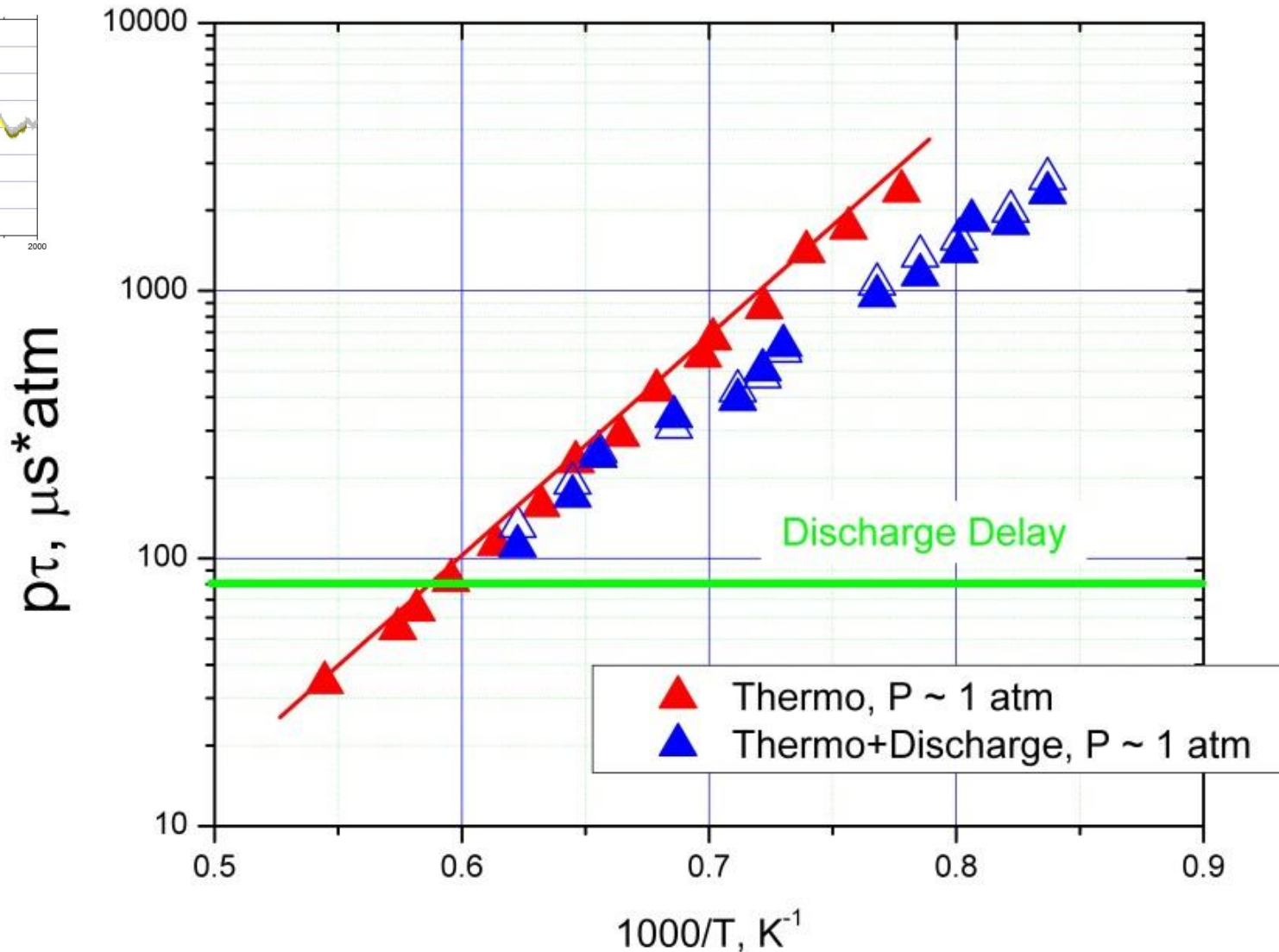
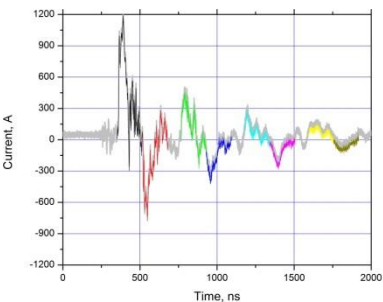
$C_2H_6:O_2:N_2:Ar = 2:7:28:63$



Combustion model: Konnov (2005)

Ignition Delay Time

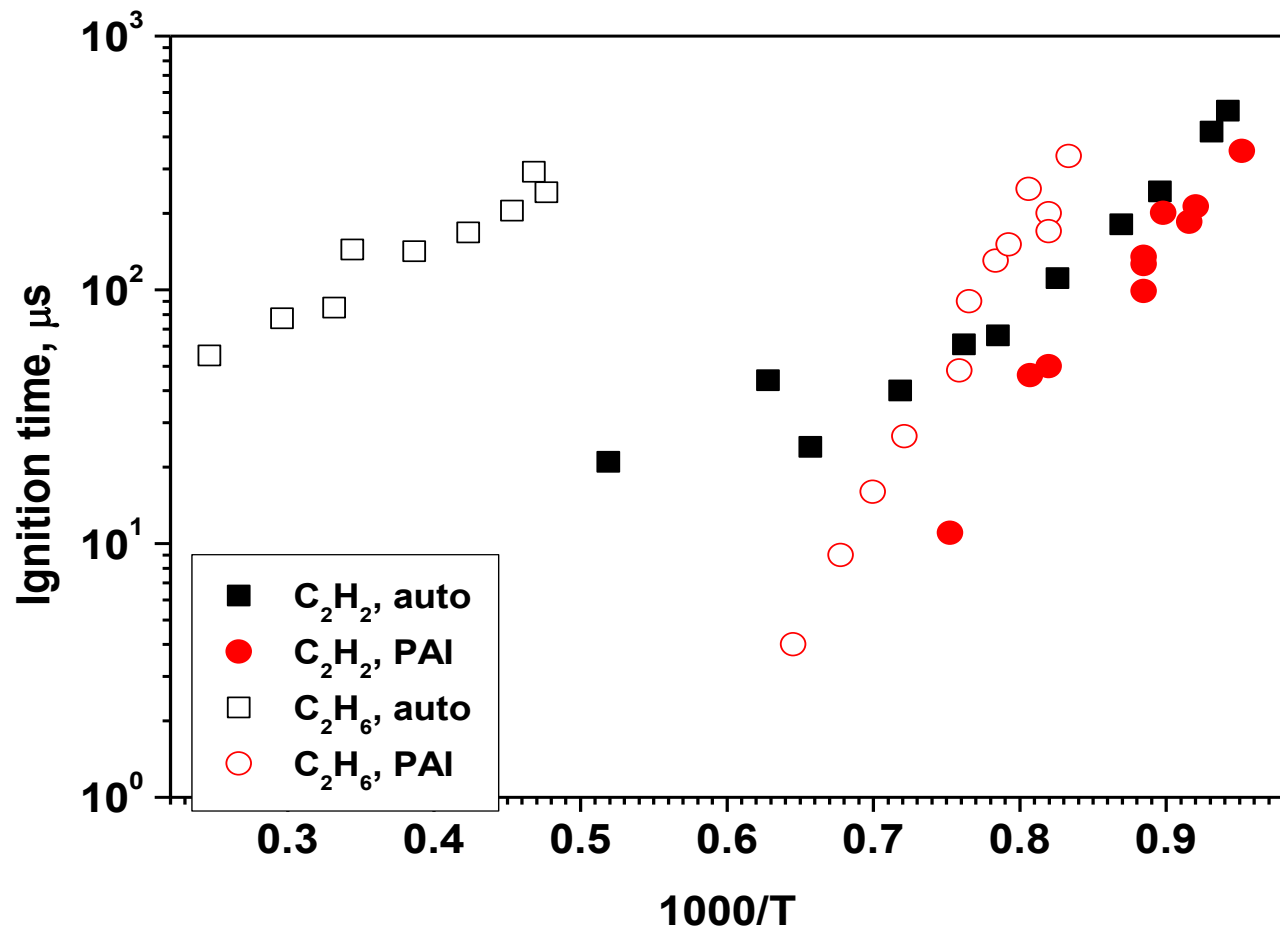
$C_2H_6:O_2:N_2:Ar = 2:7:28:63$



Combustion model: Konnov (2005)

Measured Ignition Delay Time in Stoichiometric

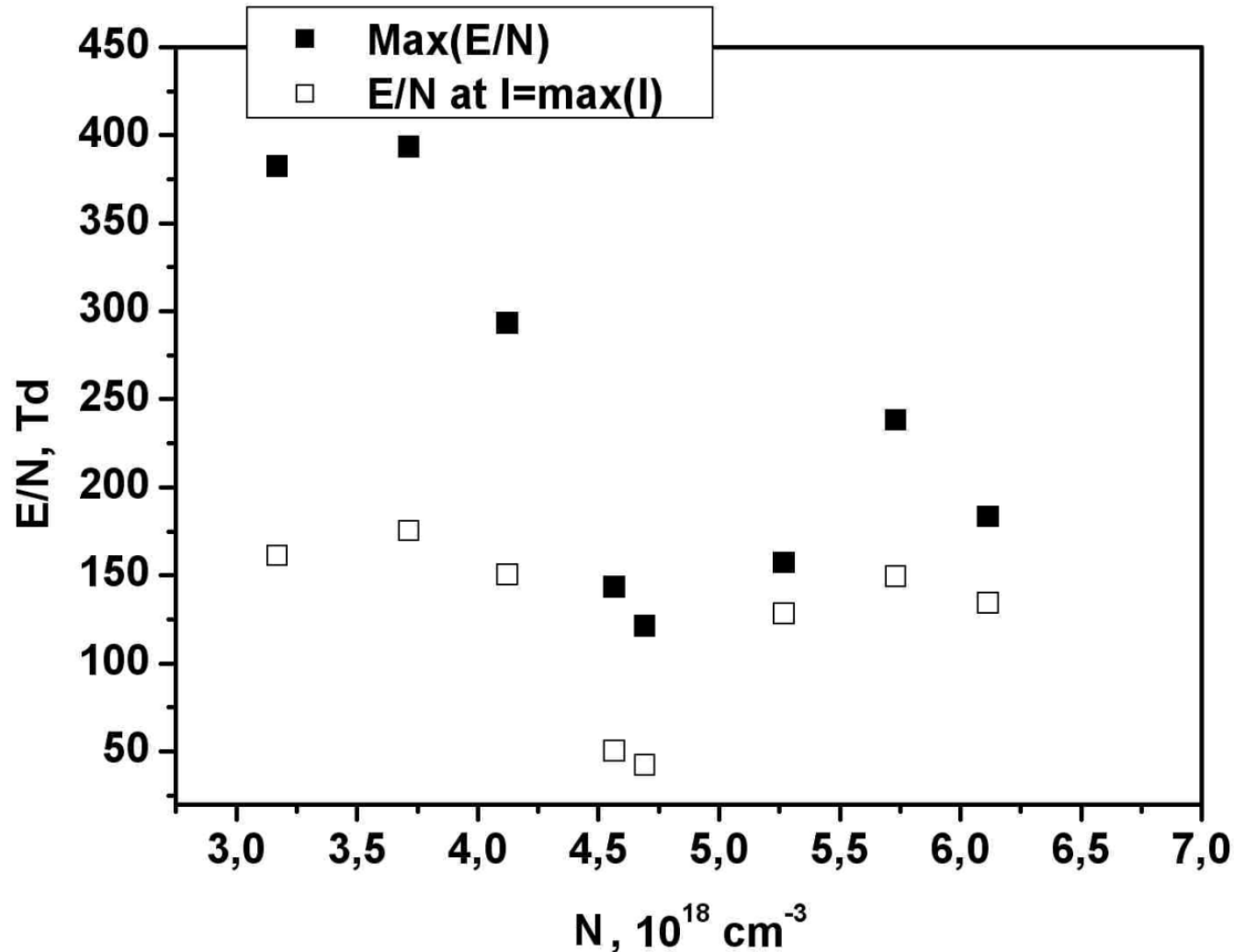
$C_2H_6:O_2:Ar$ and $C_2H_2:O_2:Ar$ Mixtures



$C_2H_6:O_2:Ar$
Kosarev et al. (2009)

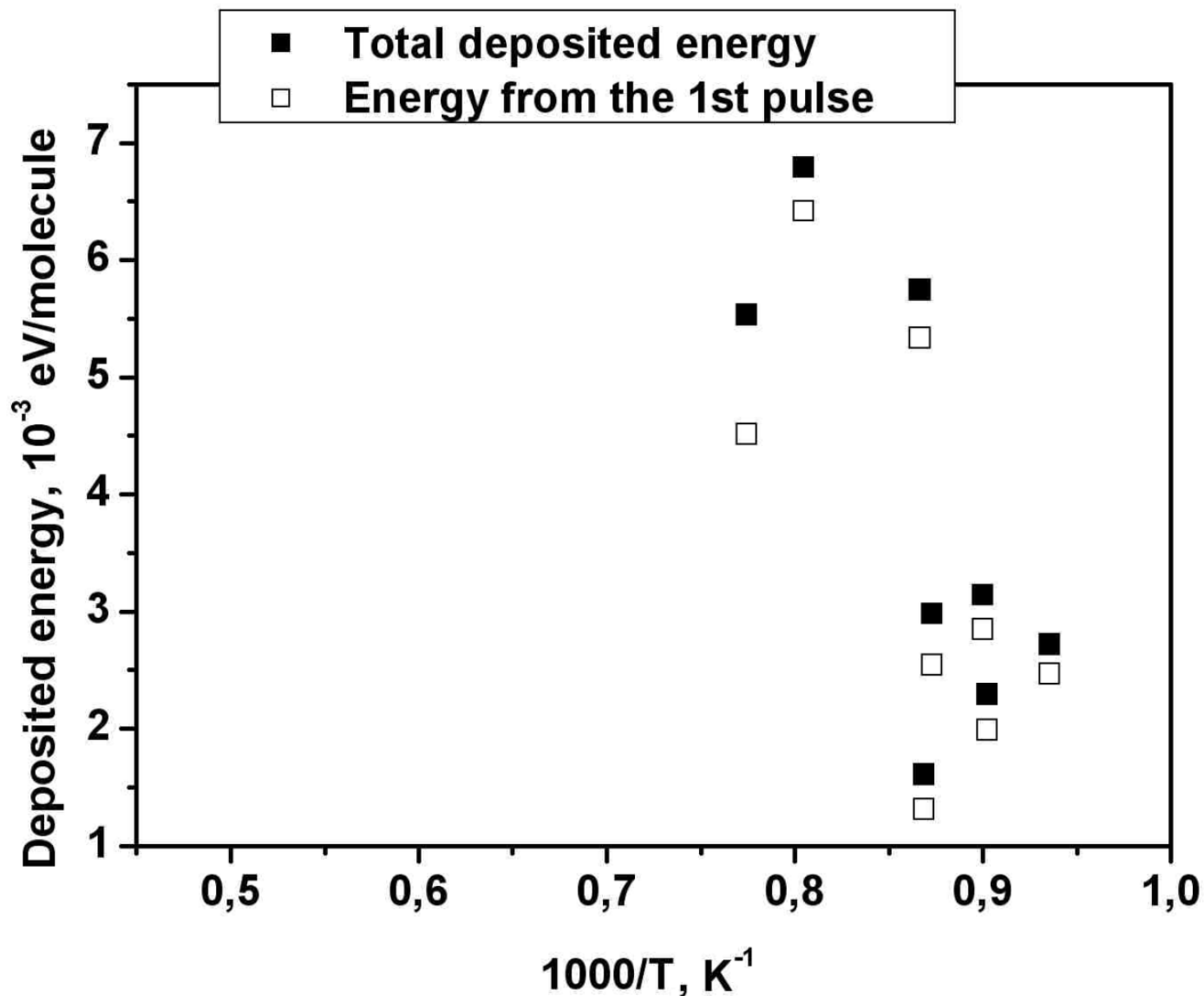
$C_2H_2:O_2:Ar$
current work

Peak Reduced Electric Field and Field at the Instant of Peak Current



**C₂H₂:O₂:Ar =
17:83:900
($\varphi = 0.5$)**

Total Specific Deposited Discharge Energy and Energy Deposited in First Pulse



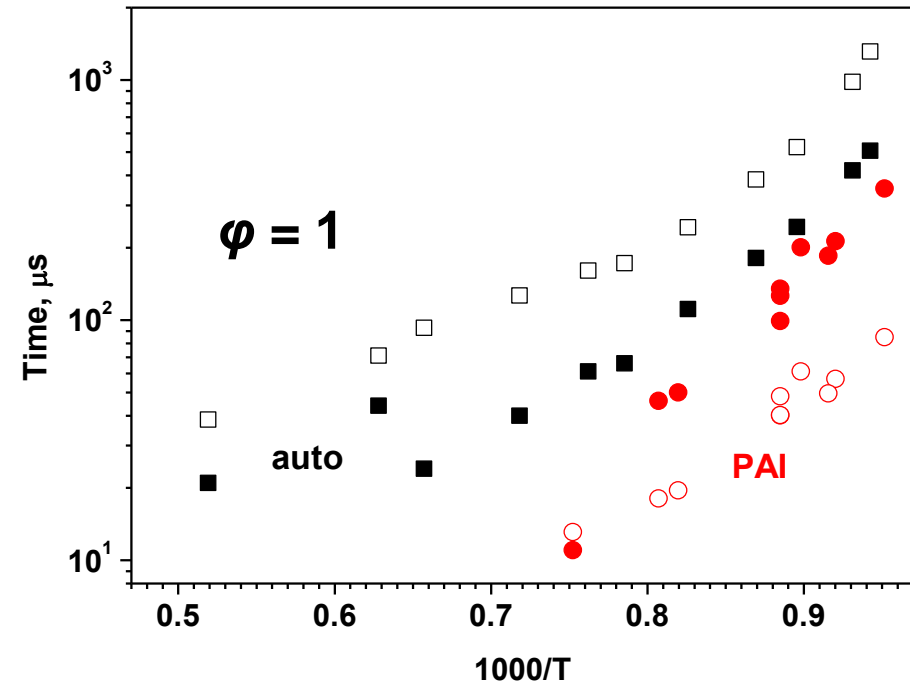
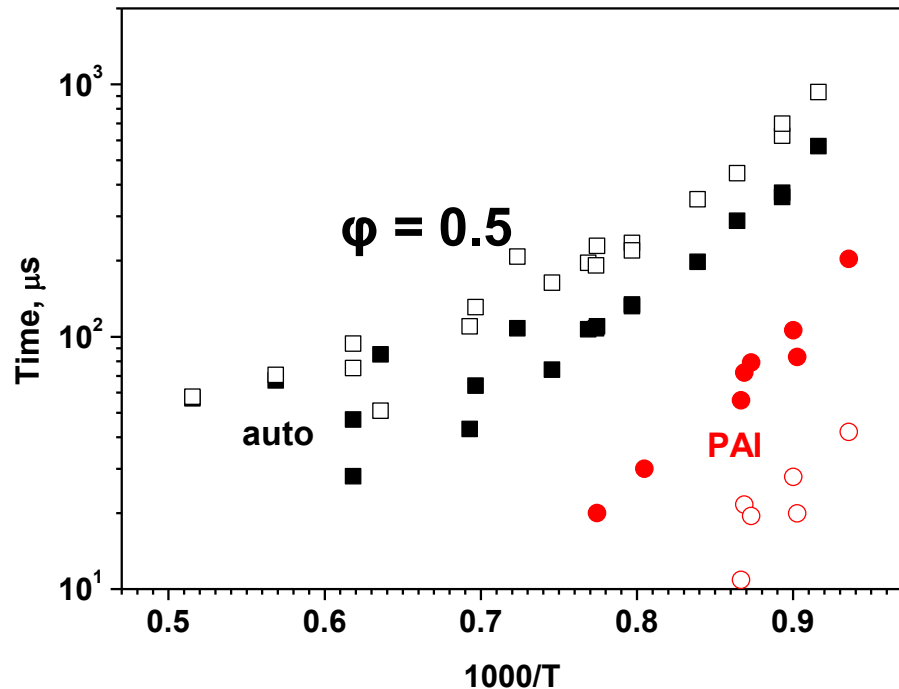
**C₂H₂:O₂:Ar =
17:83:900
($\varphi = 0.5$)**

Ignition delay time in $C_2H_2:O_2:Ar$ mixtures

solid symbols: measurements

hollow symbols: calculations with kinetic scheme

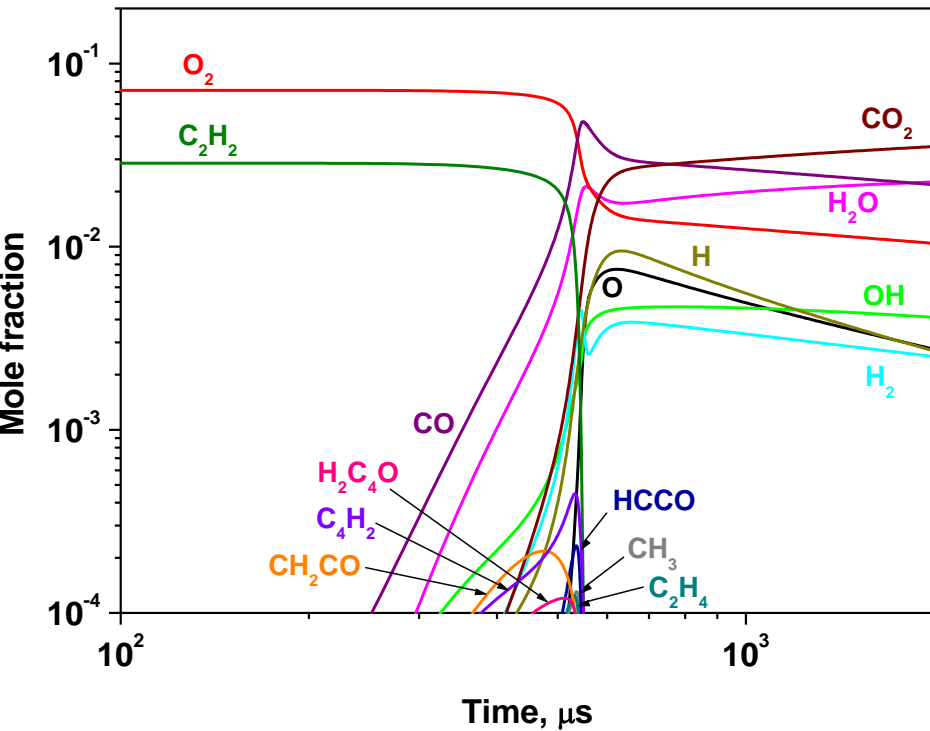
by Wang et al. (2007)



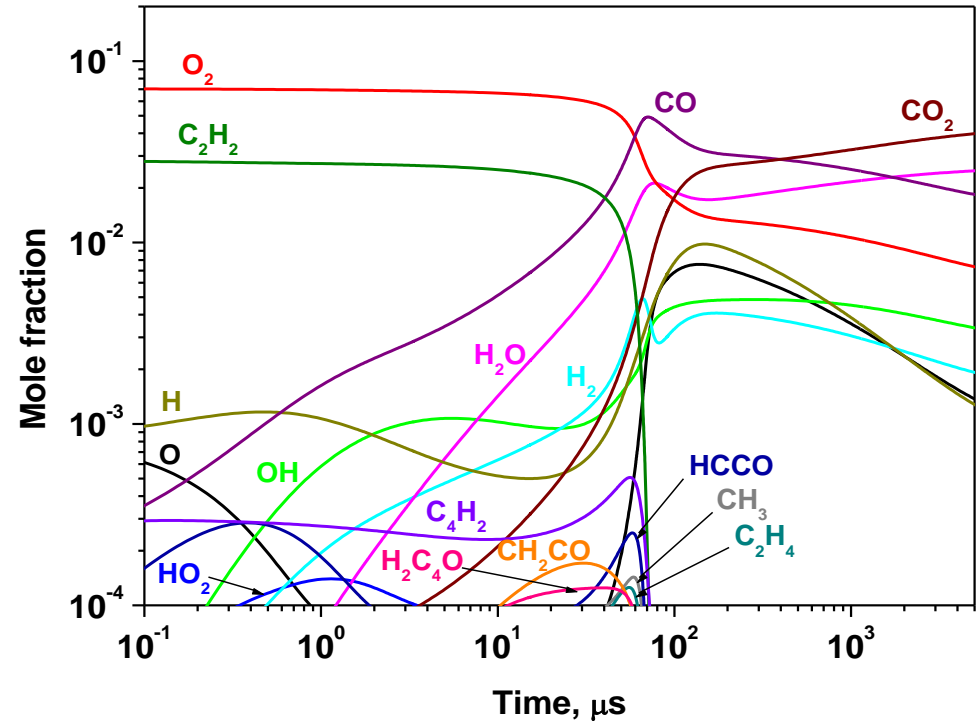
Evolution in time of calculated mole fractions for main components

Stoichiometric $C_2H_2:O_2:Ar$ mixture

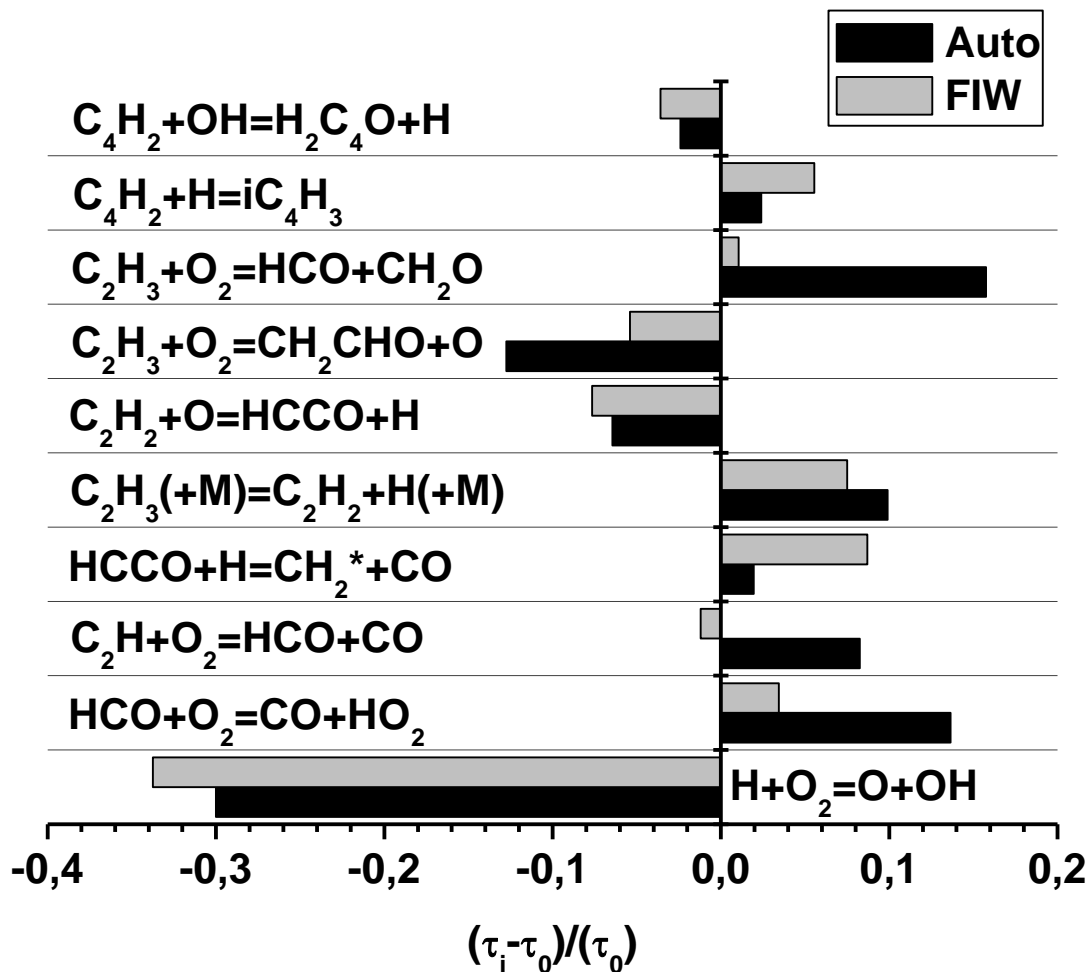
Autoignition
at 1115 K and 0.91 atm



Ignition after discharge at
1130 K and 0.91 atm



Sensitivity analysis for autoignition and ignition by discharge



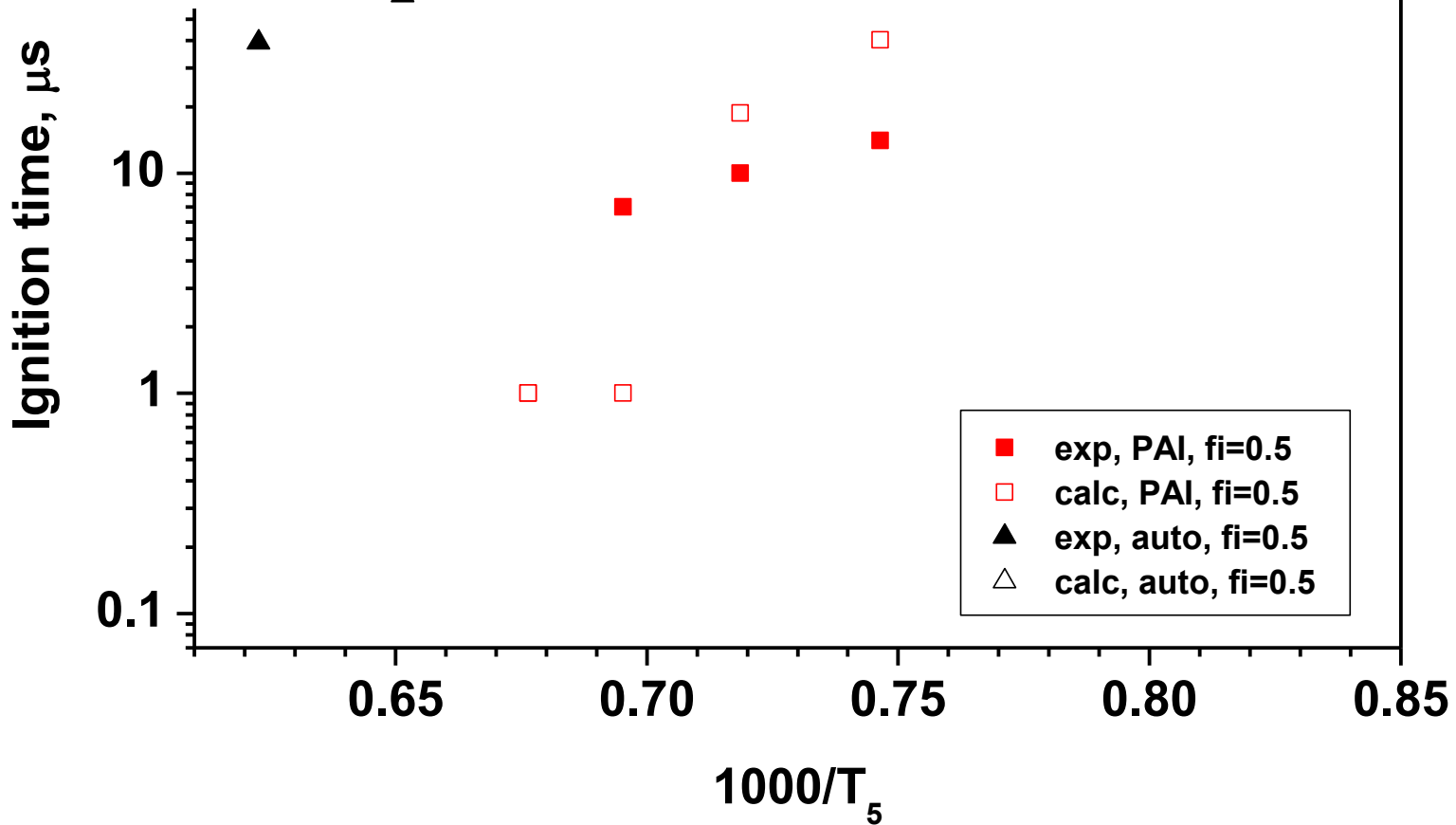
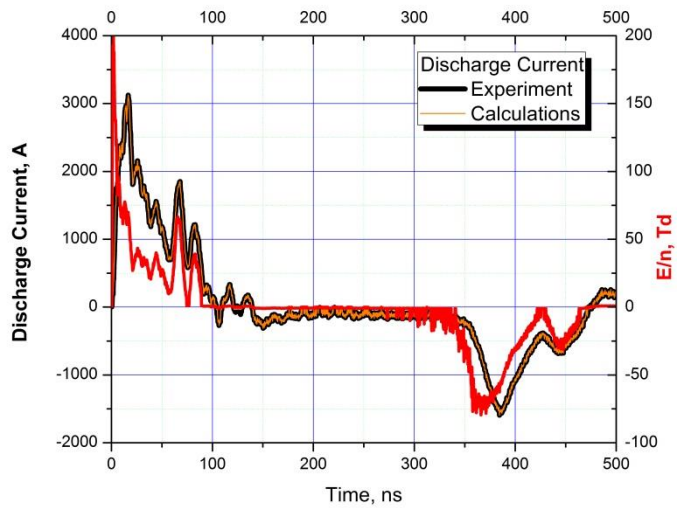
Stoichiometric
C₂H₂:O₂:Ar mixture

Autoignition
1115 K and 0.91 atm

Ignition by discharge
1130 K and 0.91 atm

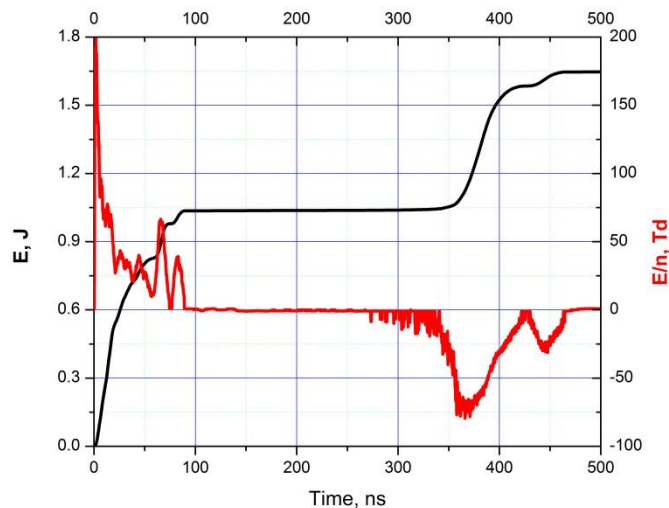
Ignition Delay Time, $\phi = 0.5$

$C_2H_5OH:O_2:Ar(90\%)$

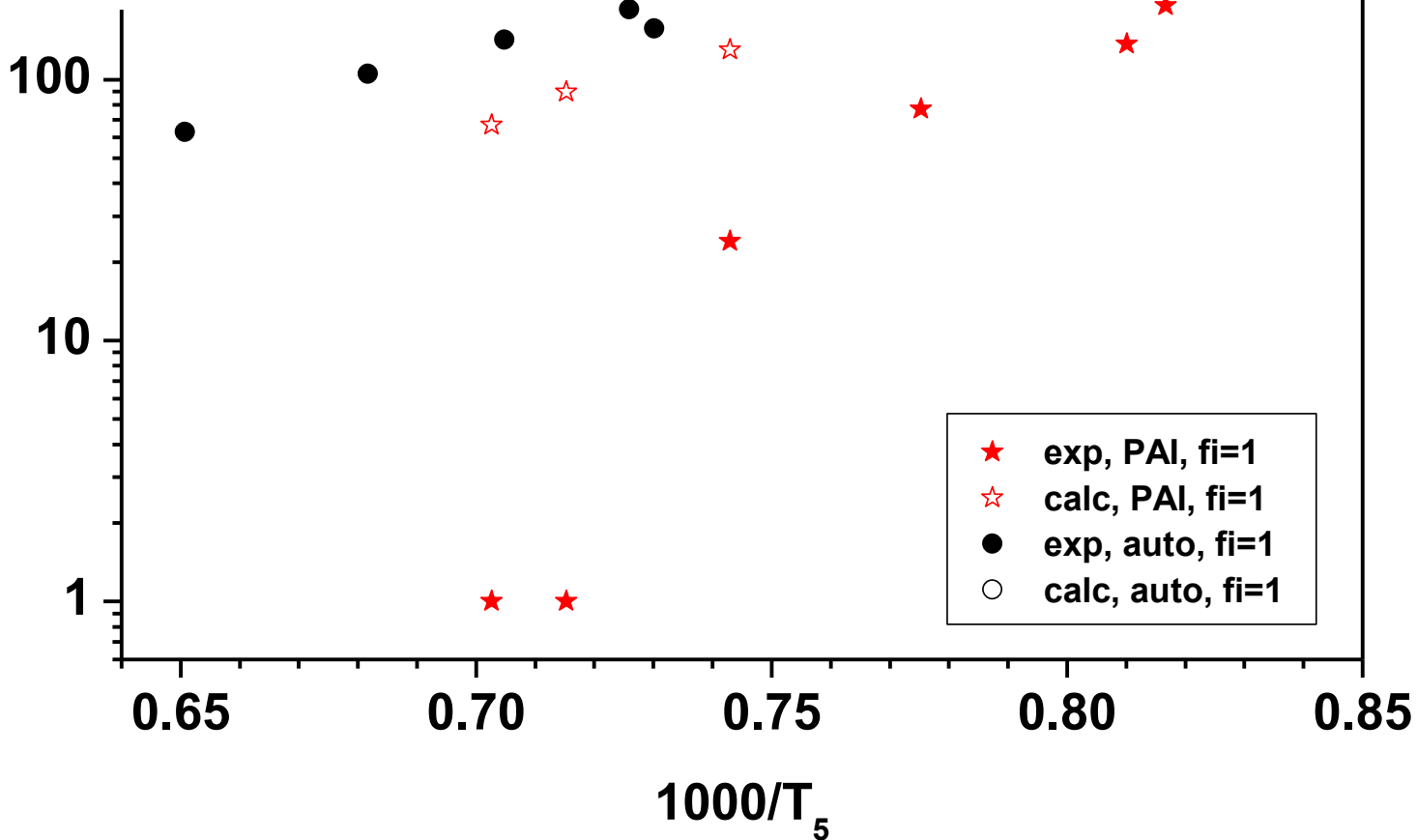


Ignition Delay Time, $\phi = 1.0$

$C_2H_5OH:O_2:Ar(90\%)$



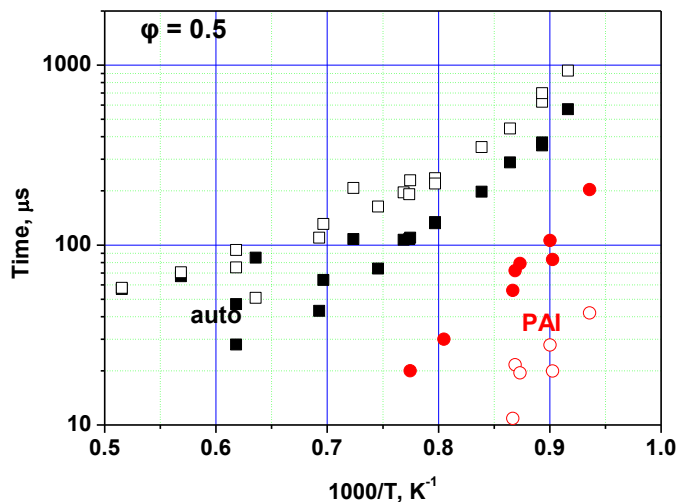
Ignition time, μs



Plasma Shock Tube Experiments Summary

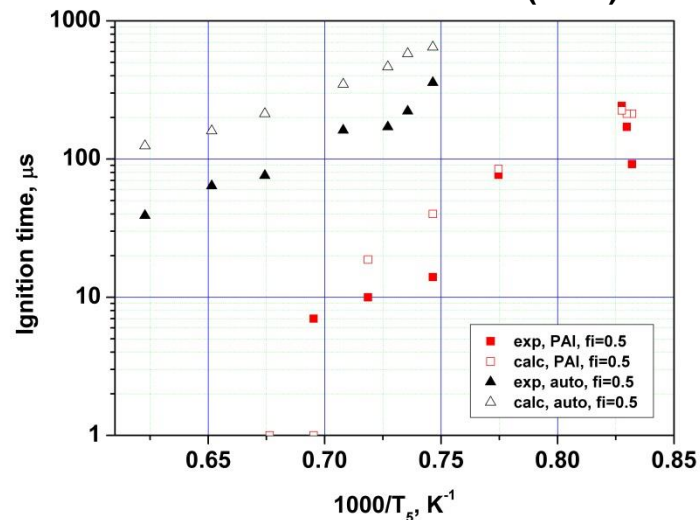
$C_2H_2:O_2:Ar(90\%)$

Combustion model: Wang et al. (2007)



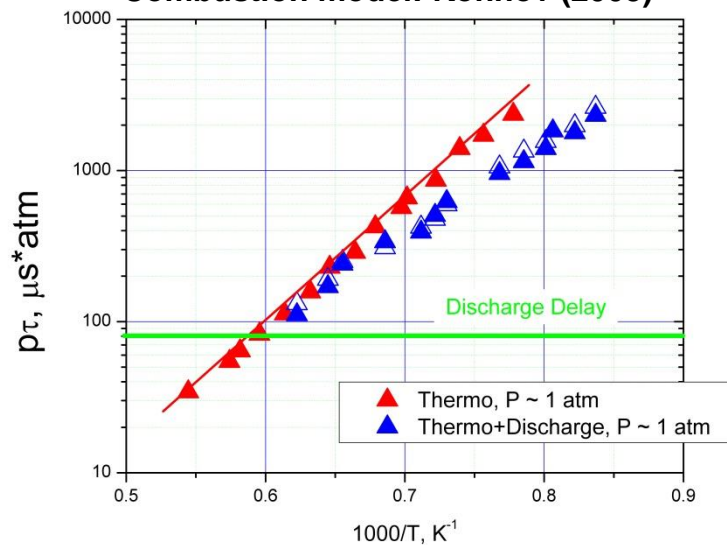
$C_2H_5OH:O_2:Ar(90\%)$

Combustion model: MARINOV (1998)



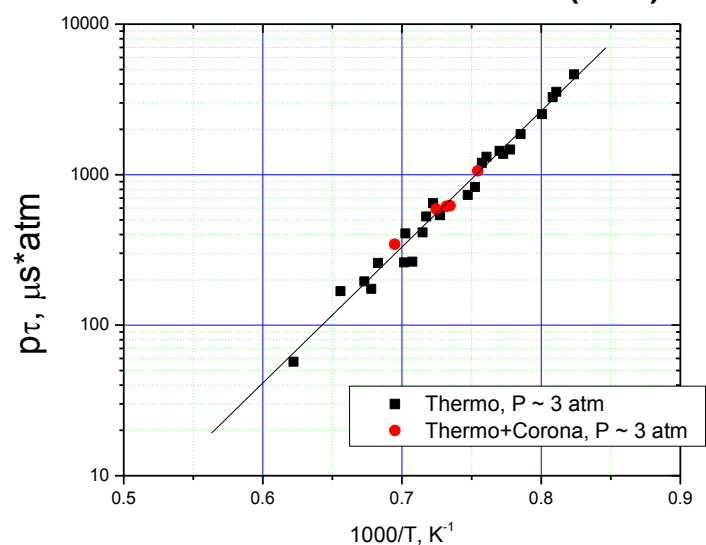
$C_2H_6:Air:Ar(63\%)$

Combustion model: Konnov (2005)

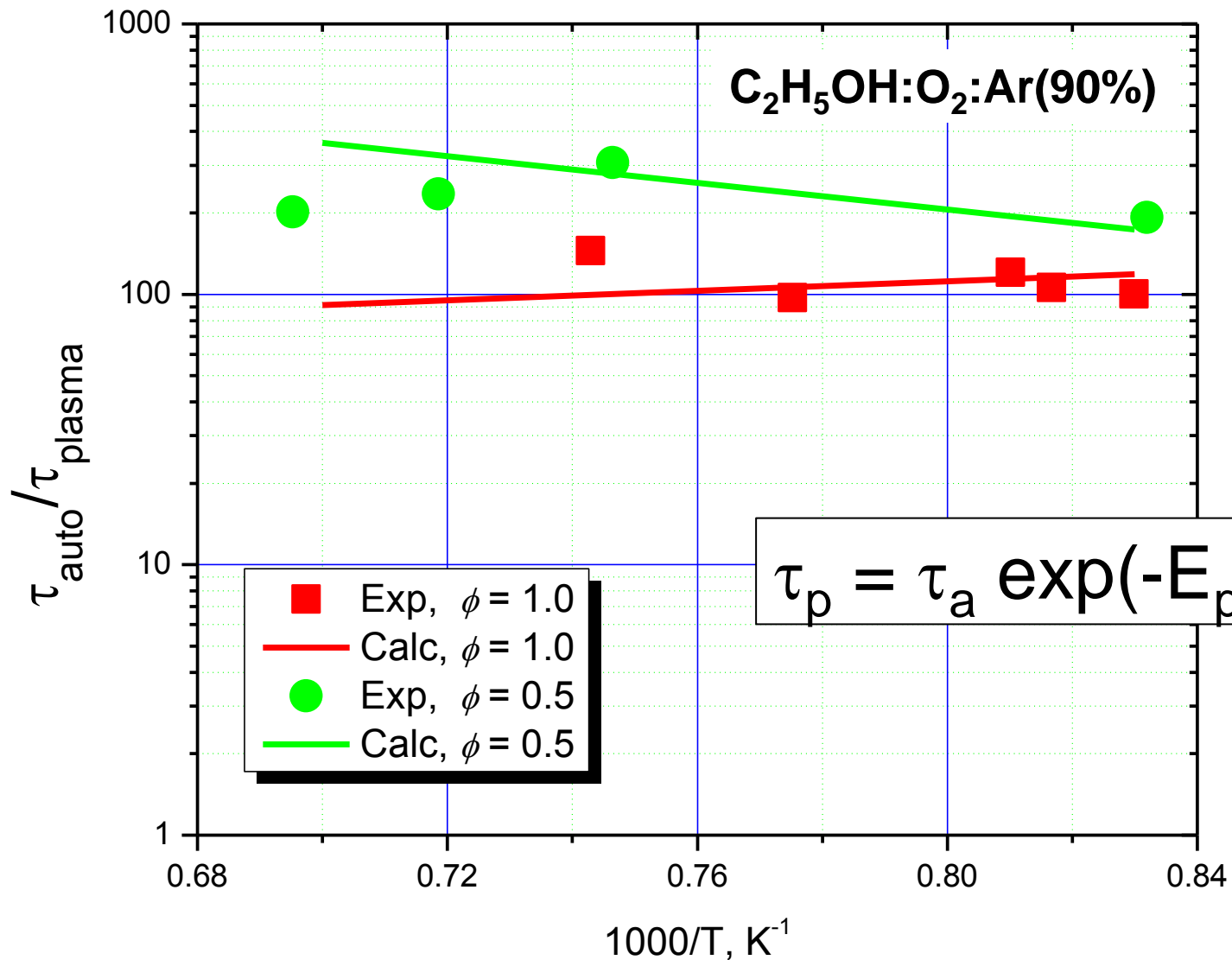


$C_2H_6:Air:Ar(63\%)$

Combustion model: Konnov (2005)

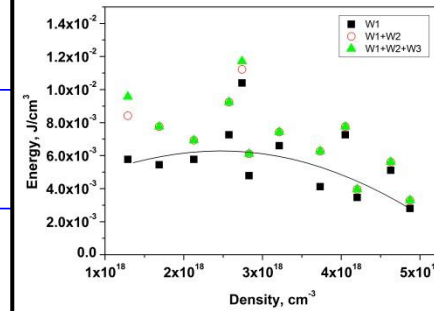
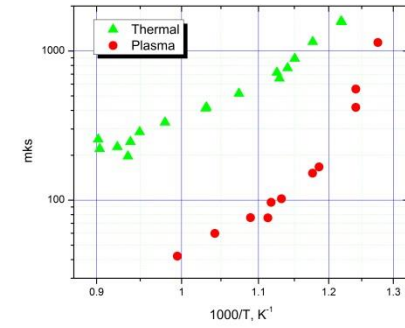
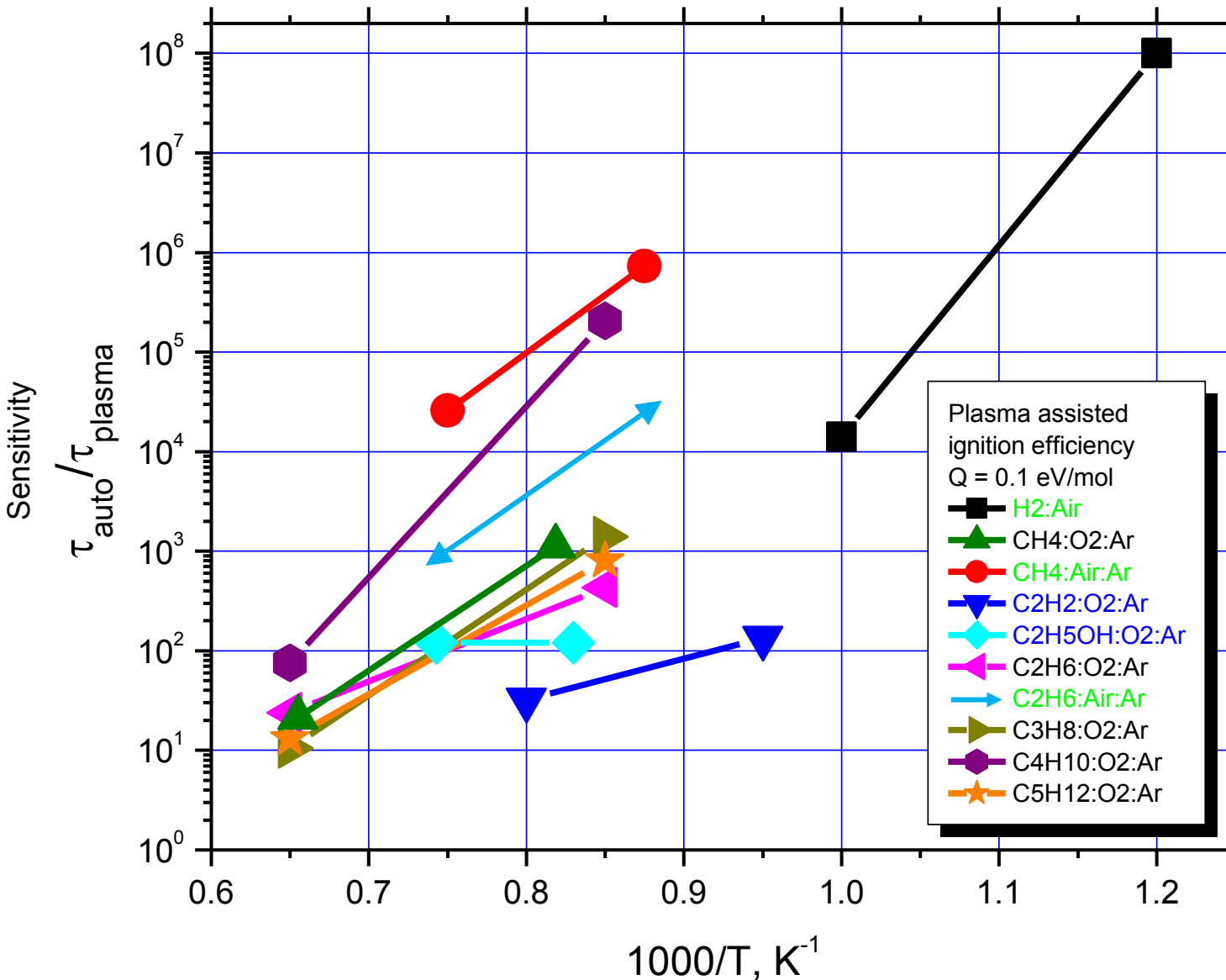


Ignition Delay Time Decrease at 0.1 eV/mol



Plasma Ignition Sensitivity

0.1 eV/mol



H₂-Air PAI

$\tau_a/\tau_p \sim 8$
 $\varepsilon \sim 0.0125 \text{ eV/mol}$

$\tau_a/\tau_{0.01} \sim 5 \text{ (-40\%)}$

$\tau_a/\tau_{0.10} \sim 2 \times 10^7$

PAC Kinetics H₂ Model Development

Plasma model:

- Plasma assisted combustion models for hydrogen oxidation understood for conditions of low energy loading per molecule. It means low ionization degree - we can neglect e-e collisions and EEDF Maxwellization due to this process.
- We have complete set of cross-sections for rotational, vibrational and electronic excitation, dissociation, dissociative ionization, ionization. These cross-sections were verified both for two-term approximation of Boltzmann equation (local EEDF) and could be modified for non-local case of extremely strong electric fields (differential cross-sections are also available).

Afterglow Model:

- **Because of fast relaxation we assume $T_{tr} = T_{rot}$ for ground state.**
- We have recombination rates for ion-electron collisions, ion-ion recombination (in some cases the products are unknown). Rates of complex ions formation/decomposition are unknown for elevated temperatures - but these ions control the plasma recombination rate.
- Quenching rates of major states are available, in some cases products are unknown. Specifically we do not know the products of reactions $N_2^* + H_2 \rightarrow \dots$

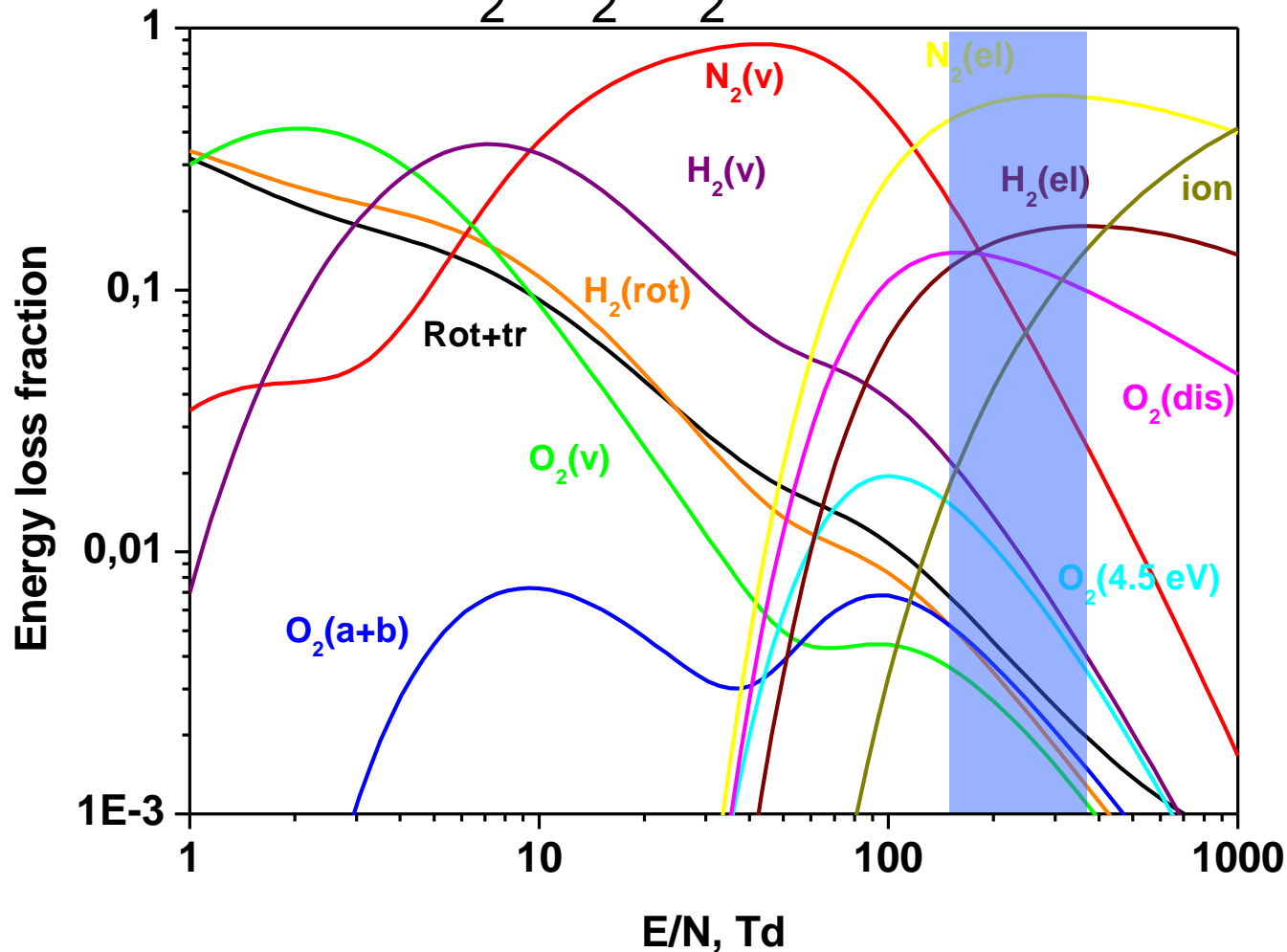
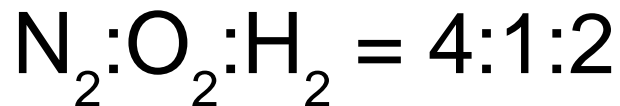
Chemical Model:

- We have complete state-to-state model of chemical reactions including vibrationally-nonequilibrium conditions for H₂-air system since 2001.
- We have verified this model for 300 K (low-P reactor), 300-800 K (1 atm streamer) and 800-1500 K (0.5 atm, reflected shock wave).

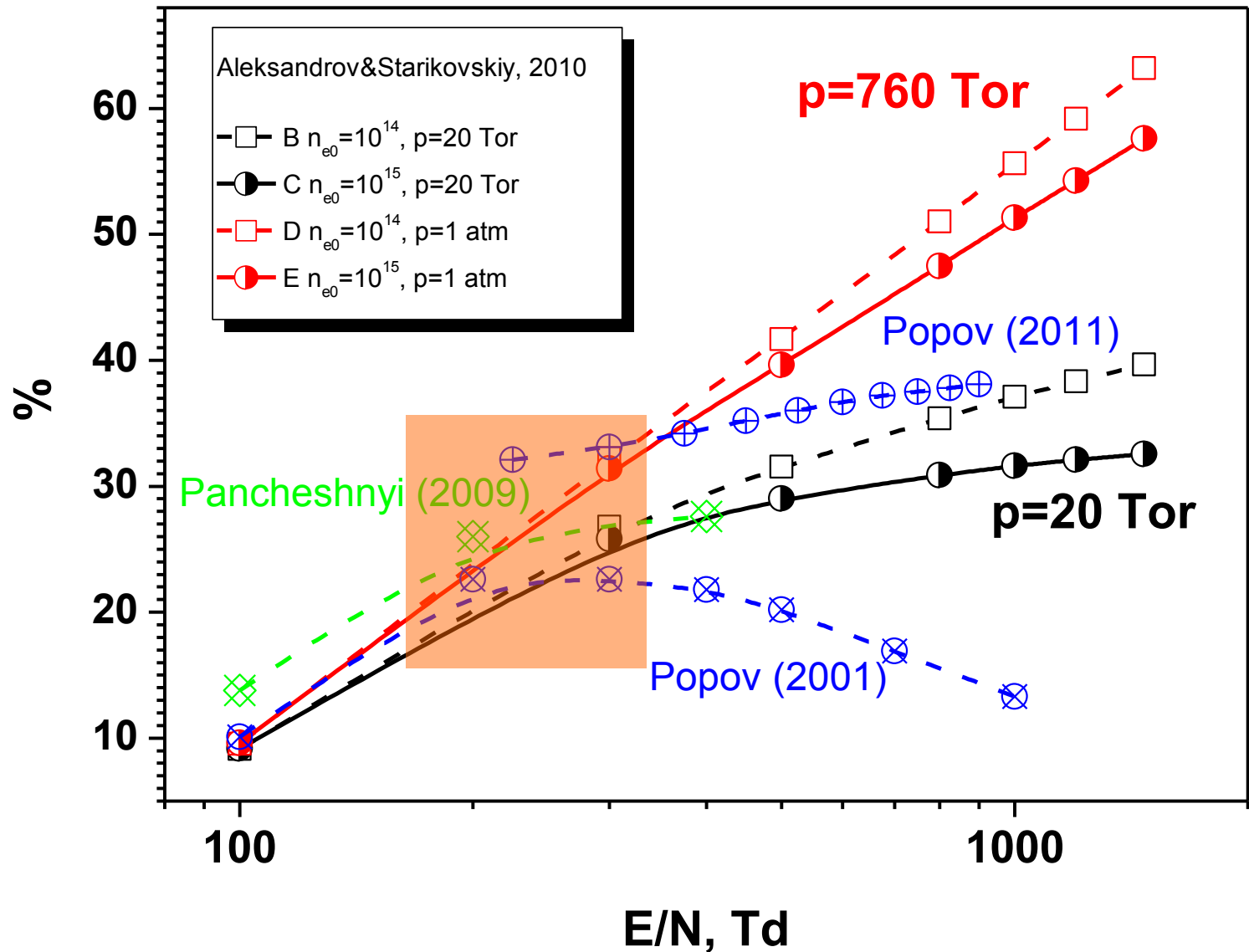
Unsolved problems:

- Because of huge number of reactions some pathways are still questionable. We need to investigate in more details the products of electron-ion and ion-ion recombination, products of electronic states dissociative quenching (focus on electronically-excited products formation).
- Reaction rate coefficients of electronically and vibrationally excited species should be verified in some cases.
- We need additional analysis of the role of complex ions in recombination and chemistry at low-T conditions.

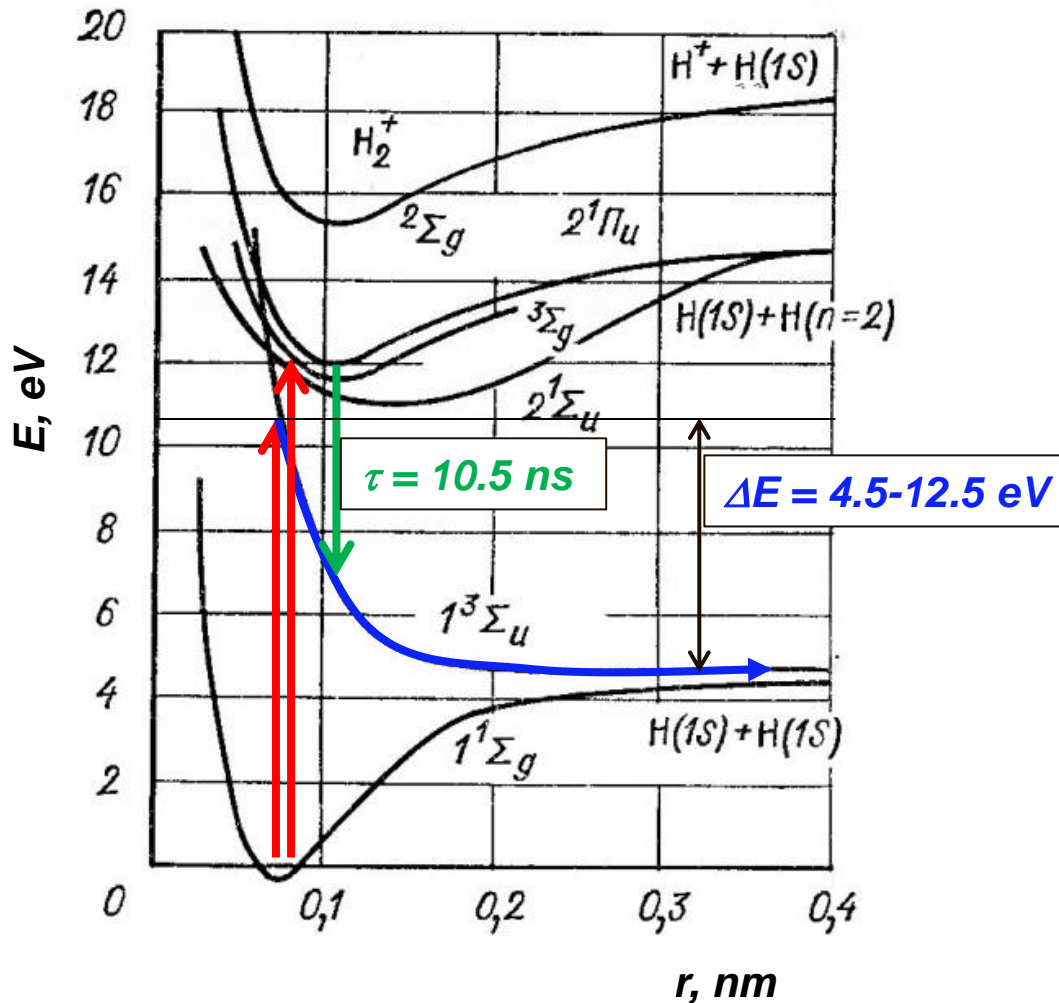
Plasma Assisted Combustion: Translational Nonequilibrium



Mechanism of Fast Heating in Air Plasma



Potential Energy Curves of Molecular Hydrogen

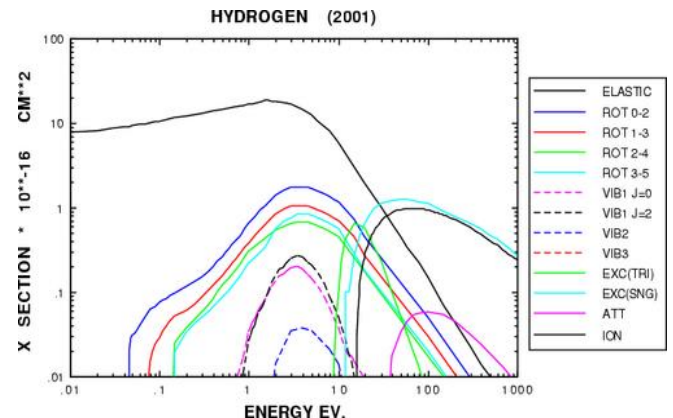


$H_2(b^3\Sigma_u)$, 8.9 eV
 $\sigma_{\text{max}} = 0.33 \text{ \AA}^2$ (17 eV)

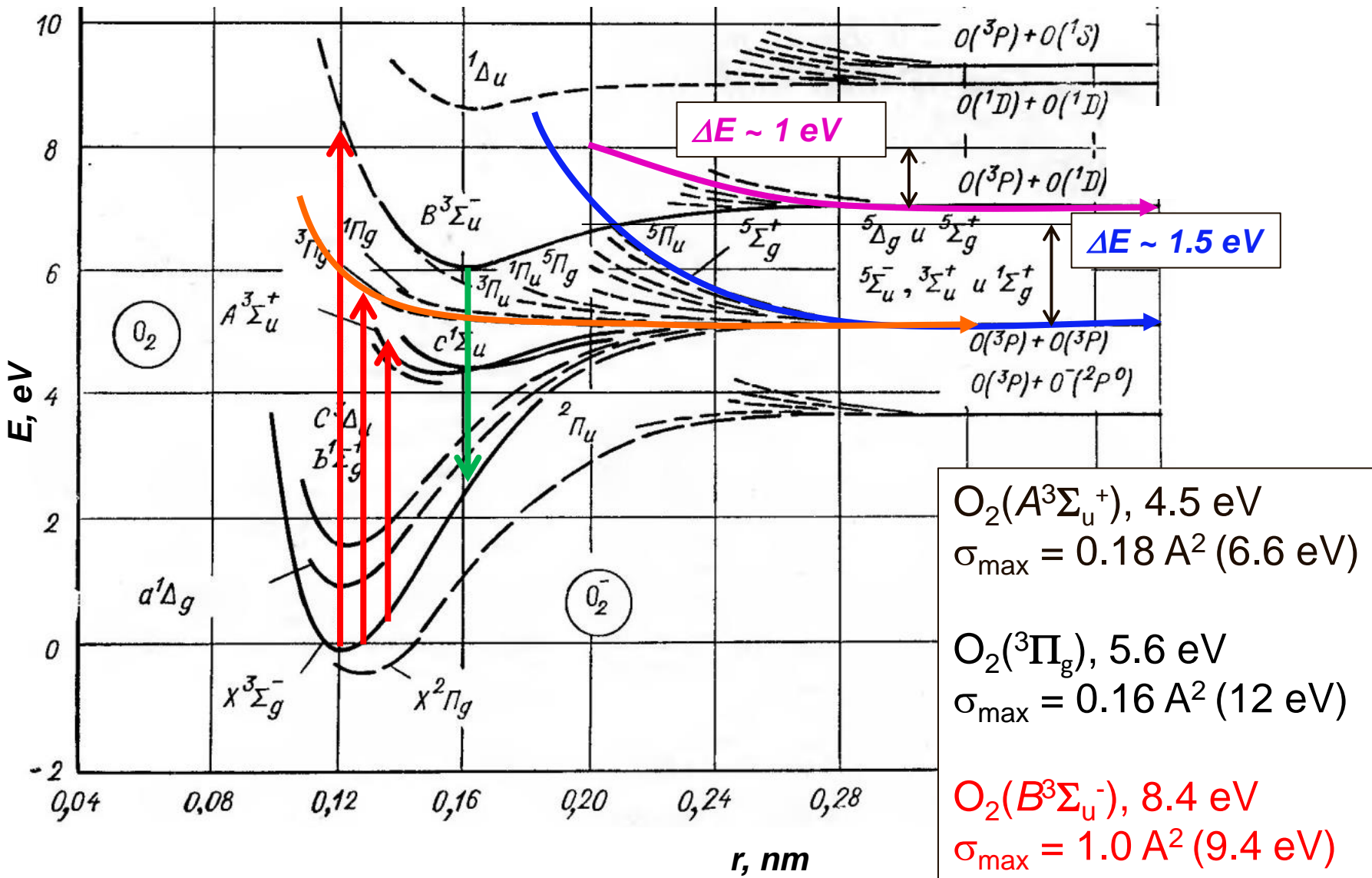
$H_2(a^3\Sigma_g)$, 11.8 eV
 $\sigma_{\text{max}} = 0.12 \text{ \AA}^2$ (15 eV)

$H_2(B^1\Sigma_u)$, 11.3 eV
 $\sigma_{\text{max}} = 0.48 \text{ \AA}^2$ (40 eV)

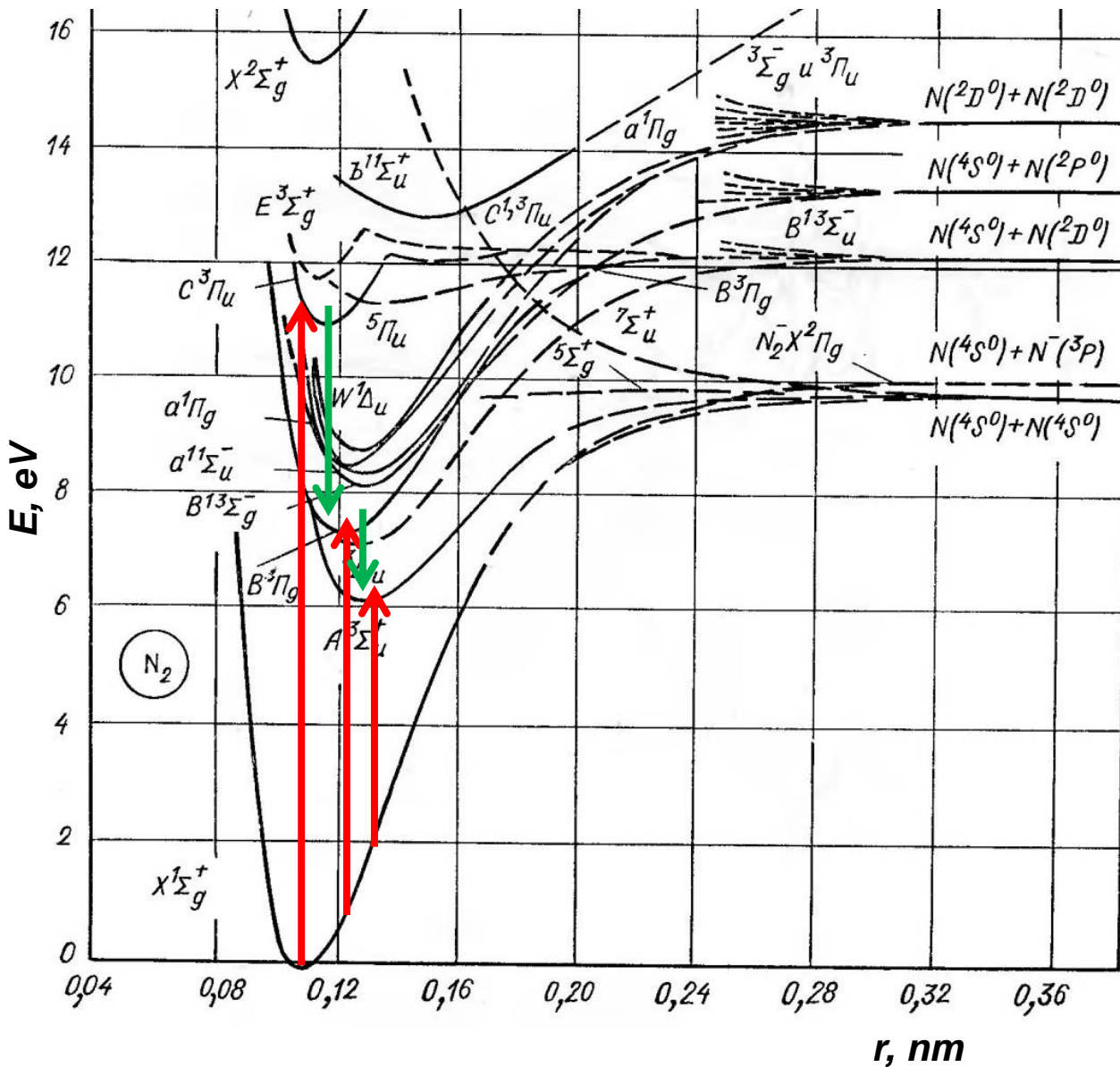
$H_2(C^1\Pi_u)$, 12.4 eV
 $\sigma_{\text{max}} = 0.40 \text{ \AA}^2$ (40 eV)



Potential Energy Curves of Molecular Oxygen



Potential Energy Curves of Molecular Nitrogen

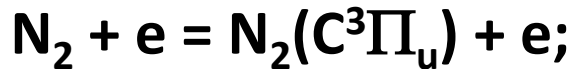


$N_2(A^3\Sigma_u^+)$, 6.2 eV
 $\sigma_{\max} = 0.08 \text{ \AA}^2$ (10 eV)

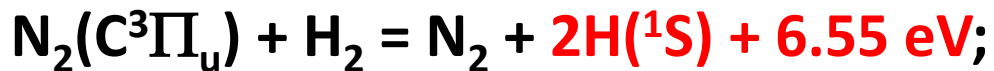
$N_2(B^3\Pi_g)$, 7.35 eV
 $\sigma_{\max} = 0.20 \text{ \AA}^2$ (12 eV)

$N_2(C^3\Pi_u)$, 11.03 eV
 $\sigma_{\max} = 0.98 \text{ \AA}^2$ (14 eV)

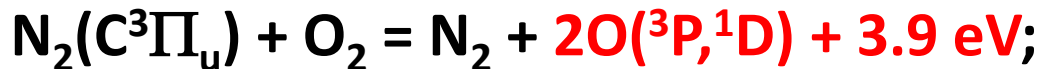
Major Channels of Hot Atoms Production



$$k = f(E/n)$$



$$k = 3.2 \times 10^{-10} \text{ cm}^3/\text{s}$$



$$k = 2.7 \times 10^{-10} \text{ cm}^3/\text{s}$$

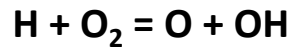


$$k = f(E/n)$$



$$k = f(E/n)$$

Chain Initiation/Branching Reactions



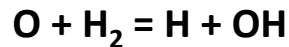
$$k = 1.6 \times 10^{-10} \times \exp(-7470/T) \text{ cm}^3/\text{s}$$

$$k(300) = 2.5 \times 10^{-21} \text{ cm}^3/\text{s}$$

$$k(\text{hot}) = 1.6 \times 10^{-10} \text{ cm}^3/\text{s}$$



$$k(300, 1 \text{ atm}) = 1.6 \times 10^{-12} \text{ cm}^3/\text{s} \quad T_{\text{crit}} \sim T_{\text{autoignition}}$$



$$k = 8.5 \times 10^{-20} \times T^{2.67} \times \exp(-3160/T) \text{ cm}^3/\text{s}$$

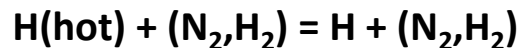
$$k(300) = 9.3 \times 10^{-18} \text{ cm}^3/\text{s}$$

$$k(\text{hot}) = 1.5 \times 10^{-10} \text{ cm}^3/\text{s}$$

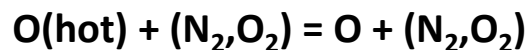
$$k(^1\text{D}) = 1.1 \times 10^{-10} \text{ cm}^3/\text{s}$$



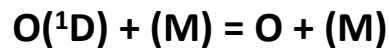
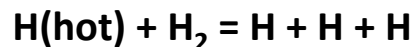
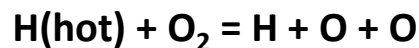
$$k(300, 1 \text{ atm}) = 2.2 \times 10^{-14} \text{ cm}^3/\text{s} \quad T_{\text{crit}} \sim 650\text{K}$$



$$k \sim 2m/M \quad k_{\text{gk}} \sim 1.6 \times 10^{-10} \text{ cm}^3/\text{s}$$



$$k \sim 2m/M \quad k_{\text{gk}} \sim 1.3 \times 10^{-10} \text{ cm}^3/\text{s}$$

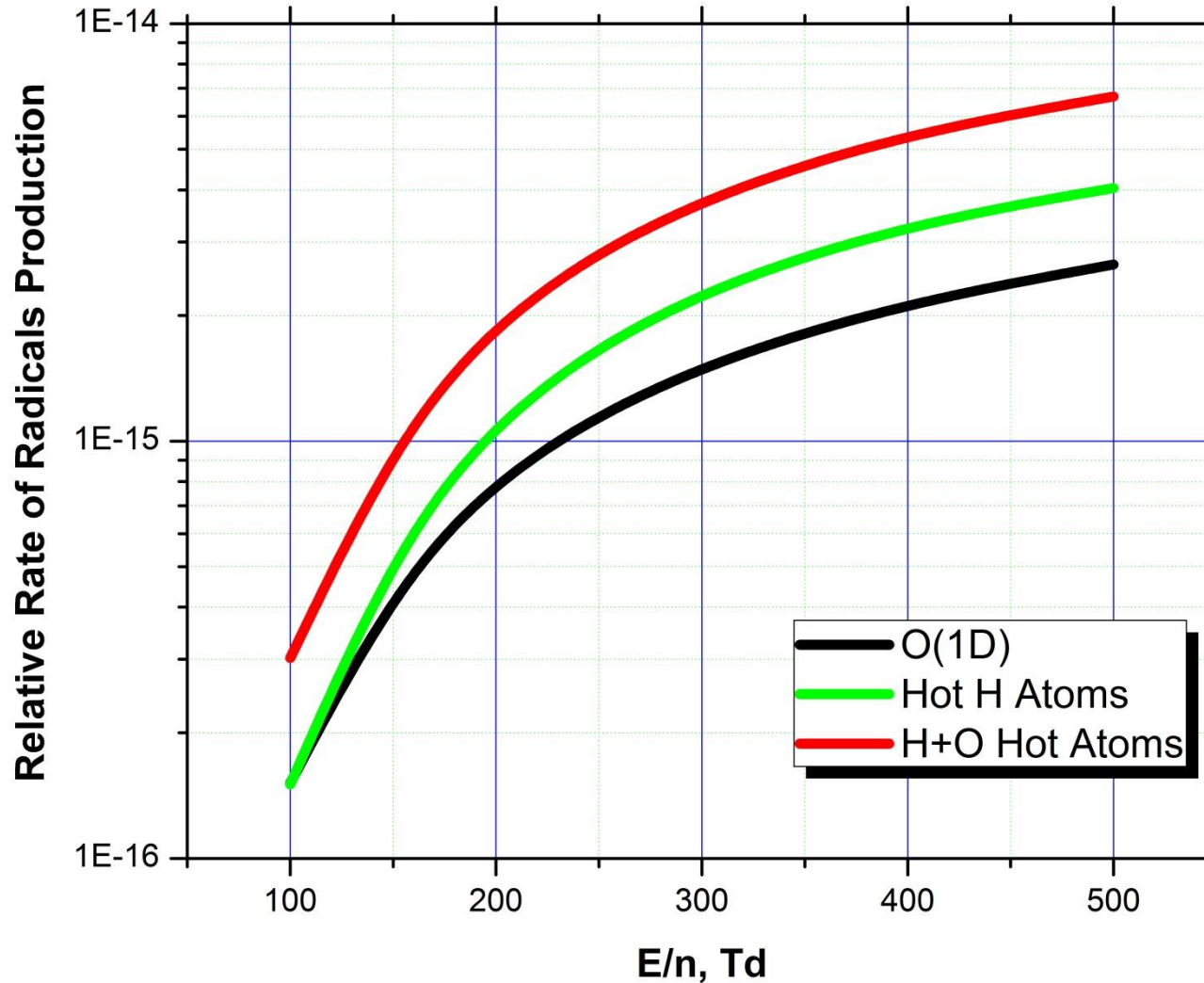


$$k = 2.6 \times 10^{-11} \text{ cm}^3/\text{s} \quad (\text{M} = \text{O}_2)$$

$$k = 1.3 \times 10^{-11} \text{ cm}^3/\text{s} \quad (\text{M} = \text{N}_2)$$

$$k = 5.2 \times 10^{-11} \text{ cm}^3/\text{s} \quad (\text{M} = \text{H}_2)$$

Radicals Production Increase in Cold H₂-Air Mixture Due to Hot Atoms Formation



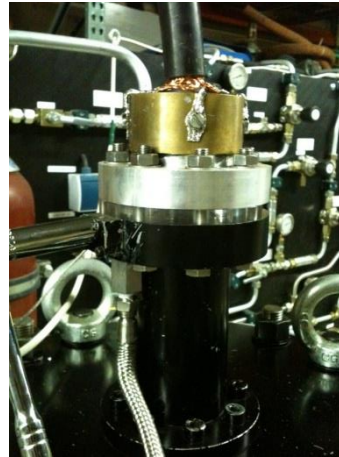
SUMMARY - 1

Experimental Facilities

1. Rapid Compression Machine.

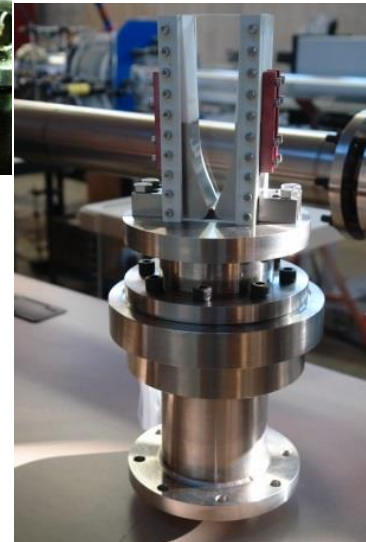
$P = 10\text{-}70\text{ atm}$, $U = 120\text{ kV}$

1 GW in 60 ns

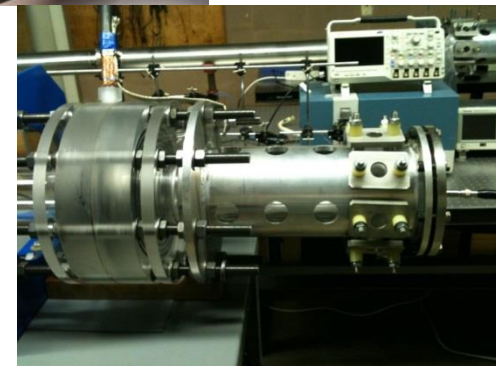


1. Plasma Shock Tunnel. $M = 3\text{-}5$, $U = 100\text{ kV}$

0.5 MW during 1 ms



1. Plasma Shock Tube. $T = 800 - 2000\text{ K}$, $U = 120\text{ kV}$

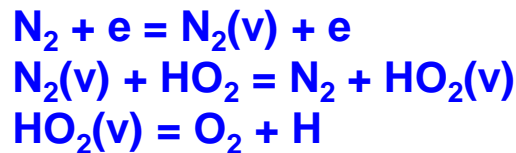
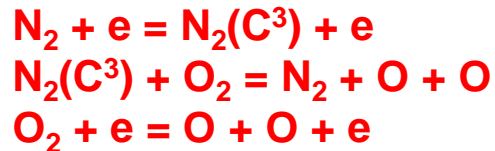


SUMMARY - 2

Major Results

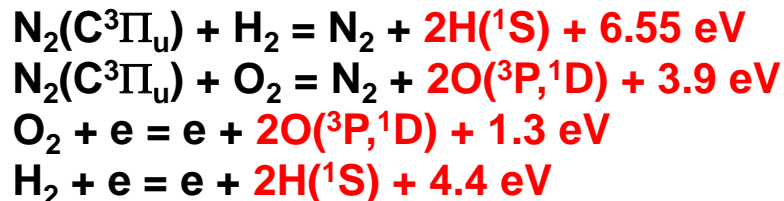
Two new mechanisms of PAC proposed:

1) Influence of Vibrational Excitation on Low-Temperature Kinetics



Synergetic Effect of High and Low Electric Fields

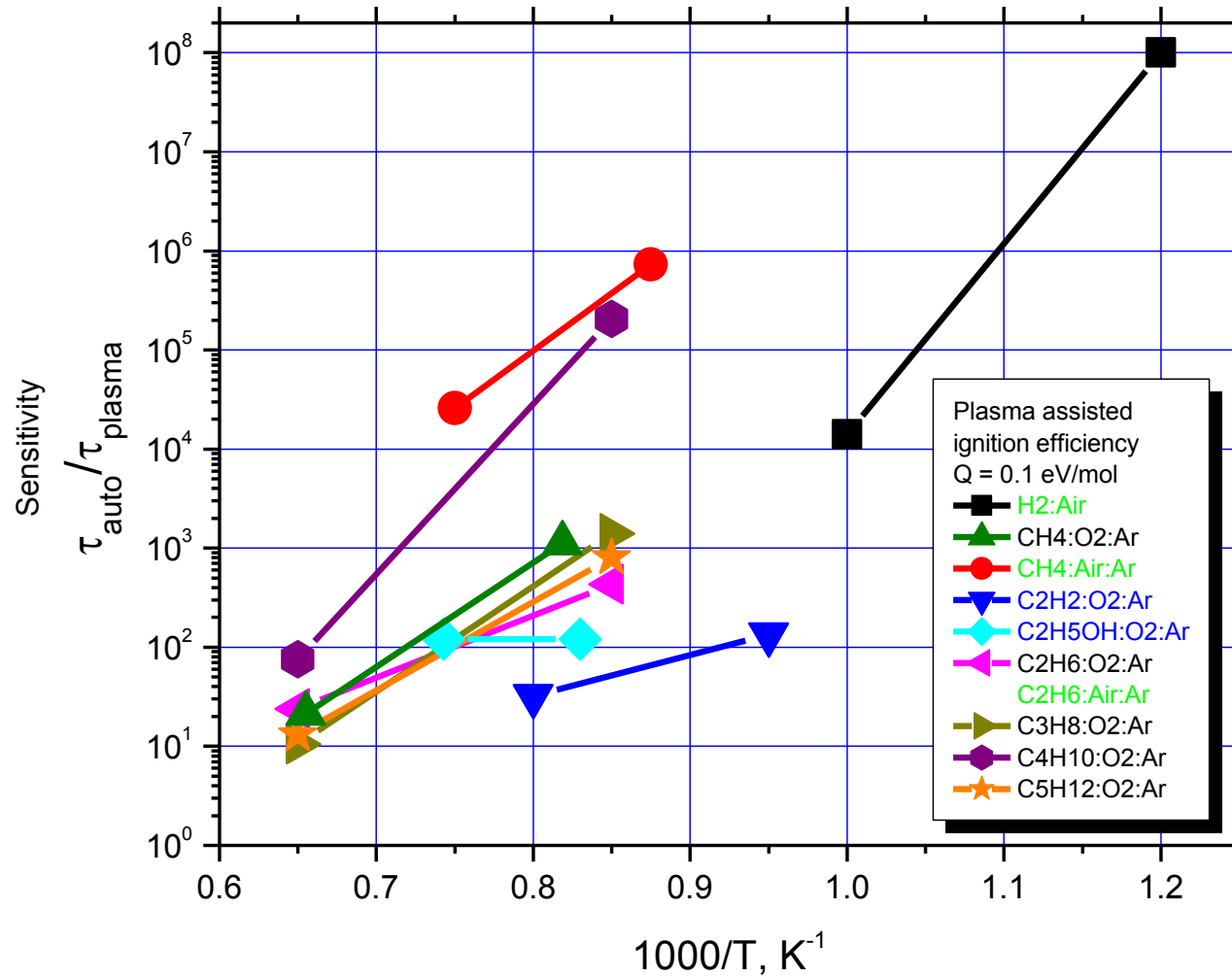
2) Radicals Production Increase Due to Hot Atoms Formation



SUMMARY - 3

Major Results

Plasma Ignition Efficiency for Different Fuels Analyzed



Future Plans

- 1) Role of Translational and Vibrational Nonequilibrium Analysis of non-Boltzmann, non-Maxwell regimes of reactions
- 2) Reference Experiments Database for PAC
High-pressure regimes (RCM)
Low-pressure regimes (STube)
High-speed conditions (STunnel)

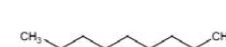
- 3) “Best Fuel for PAC”

Jet Fuel Composition

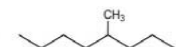
Ideal Carbon Length C8-C16

Paraffins

70%-85%



Normal Paraffins



Iso-Paraffins



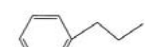
Cyclic Paraffins

Aromatics

<25%



Toluene



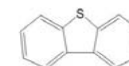
n-propylbenzene



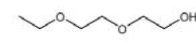
Naphthalene

Trace Compounds

<1%



Dibenzothiophene



*Diethylene glycol
monomethyl ether*



Phenol

Major International Collaborations and International Projects

Nickolay Aleksandrov (MIPT, Russia)

Ilya Kosarev (MIPT, Russia)

Sergey Pancheshnyi (ABB, Austria)

Svetlana Starikovskaya (LPP, France)

PROJECTS:

PARTNER UNIVERSITY FUND “Physics and Chemistry of Plasma-Assisted Combustion” (Princeton-LPP)

RUSSIAN FEDERAL PROGRAM “Plasma-Assisted Combustion Ultra-Lean Fuel-Air Mixtures for Energy Devices Efficiency Increase” (Princeton-MIPT)