Laminar burning velocities of stoichiometric inert-diluted methane-N_2O flames

Maria Mitu, Codina Movileanu, Venera Giurcan, Adina Magdalena Musuc, Domnina Razus

"Ilie Murgulescu" Institute of Physical Chemistry, Romanian Academy, Bucharest, Romania, 202 Spl. Independentei, 060021 Bucharest, Romania

E-mails: maria_mitu@icf.ro; cmovileanu@icf.ro; venerab@icf.ro; amusuc@icf.ro; drazus@icf.ro.

OBJECTIVES

- Study of the laminar burning velocities of stoichiometric methane-nitrous oxide gaseous mixture diluted by He, Ar, N_2 or CO_2 at various initial pressures and temperatures, by means of dynamic pressure records of explosions in a spherical vessel with central ignition.
- Study of the influence of initial pressure and additive type (He, Ar, N_2 or CO_2) on flame propagation, at 300 K.
- Calculation of the laminar burning velocities, S_u, from the constants of cubic law of pressure rise in the incipient stage of flame development.
- Examination of the laminar burning velocities obtained from experimental data against the propagation velocities obtained by the kinetic modelling of flame propagation, using COSILAB 1D package and GRI 3.0 mechanism.
- Comparison of volumetric rates of heat release, of temperature profiles across the flame front of stoichiometric CH_4-N_2O mixture diluted with 50% inert and mass fraction profiles – active species in the flame front.

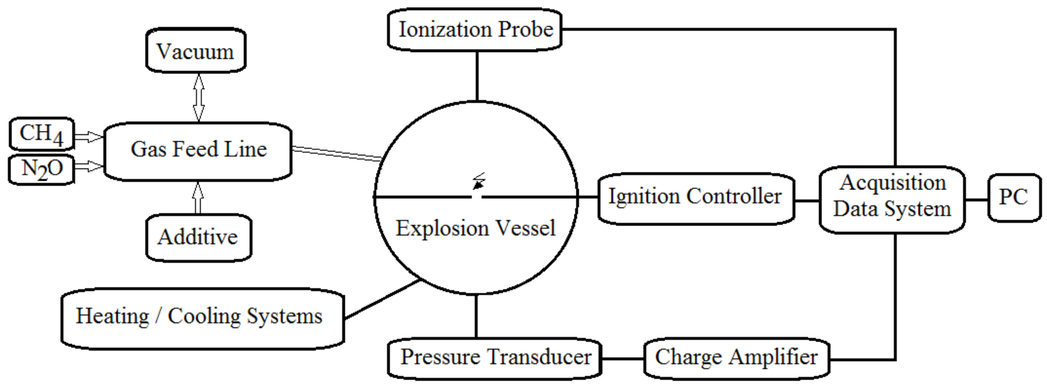
EXPERIMENTAL

- Initiation: inductive-capacitive electric sparks
- Combustion vessel: spherical, (Φ = 10 cm and V = 0.52 L), stainless steel electrodes, central position of spark gap
- Measuring systems:
  - 2 ionization probes; tips 3 mm from wall, equatorial positions
  - Pressure transducer Kistler 601 A; Charge Amplifier Kistler 5001SN
- Data acquisition: Tektronix TestLab 2505, 4 channels, 100 MHz
- Combustion vessel: spherical, (Φ = 10 cm and V = 0.52 L), stainless steel electrodes, central position of spark gap
- Experiments - made with stoichiometric methane – nitrous oxide mixtures diluted with 50% gaseous additives at various initial pressures and constant initial temperature:

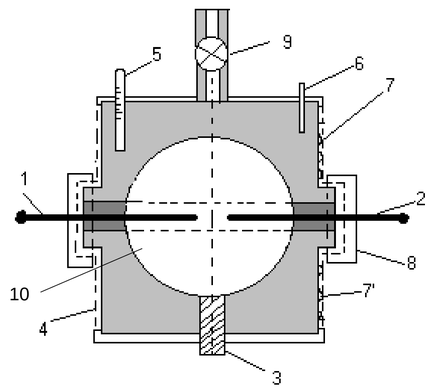
  $[CH_4]$ = 10 vol.% ; $[N_2O]$ = 40 vol.% ; [Inert] = 50 vol.% (He, Ar, N_2 or CO_2)

  p_0 = 0.50 – 1.75 bar; T_0 = 300 K

Schematic diagram of the experimental setup



Explosion vessel



1,2 electrodes
3 pressure transducer
4 thermal isolation
5 thermometer
6 thermocouple
7,7' heating resistors
8 isolation layer
9 admission / evacuation valve
10 ionization probes

Data evaluation

The laminar burning velocity, S_u, of gaseous flammable mixtures at initial pressure p_0 was calculated in the early stage of the flame propagation as [1,2]:

$$S_u = R \cdot \left(\frac{k}{\Delta p_{max}}\right)^{1/3} \cdot \left(\frac{p_0}{p_{max}}\right)^{2/3}$$

where R is the vessel's radius, k is the coefficient of the cubic law of pressure rise, Δp_{max} is the maximum (peak) pressure rise of the explosion and $p_{max} = p_0 + \Delta p_{max}$.

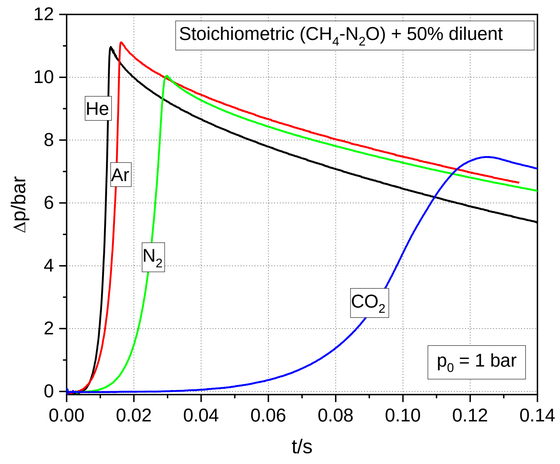
The cubic law coefficient was determined for each experiment by a nonlinear regression of the form [1,2]: $\Delta p = a + k \cdot (t - b)^3$, where a and b are pressure and time corrections, respectively.

COMPUTER PROGRAM

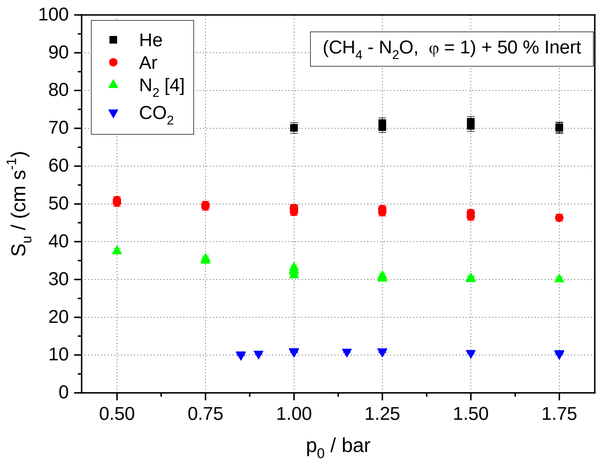
The kinetic modelling of the laminar adiabatic premixed flames, in various conditions, was made with COSILAB-1D package [3] which delivered the laminar burning velocity, S_u, and the profiles of temperature and chemical species present in the flame.

In the present case the GRI 3.0 mechanism was used, and 53 chemical species and 325 elementary reactions were involved. The thermodynamic and molecular databases (format for CHEMKIN) of Sandia National Laboratories, USA, provided the input data for the runs.

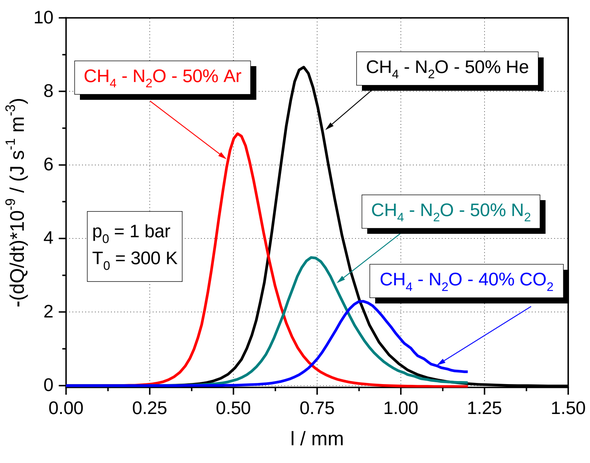
RESULTS



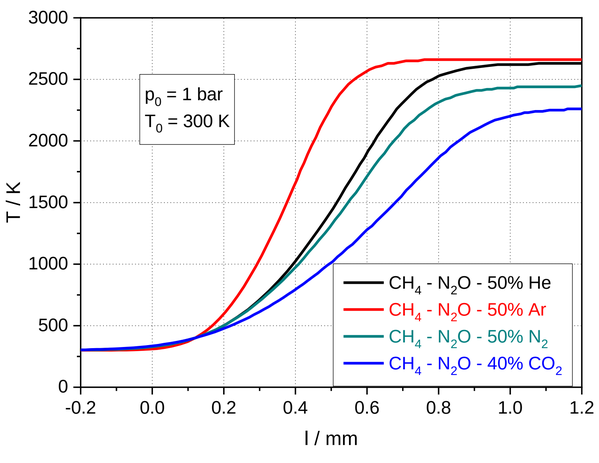
Pressure-time records of explosions in the stoichiometric CH_4-N_2O mixture diluted with 50% inert (He, Ar, N_2 or CO_2) at p_0 = 1.0 bar, and T_0 = 300 K.



Pressure influence on experimental burning velocities of 50 % inert-diluted stoichiometric CH_4–N_2O mixture



Volumetric rates of heat release across the flame of stoichiometric CH_4-N_2O mixture diluted with inert (50% He, Ar, N_2 and 40% CO_2)



Temperature profiles across the flame front of the stoichiometric CH_4-N_2O mixture diluted with inert (50% He, Ar, N_2 and 40% CO_2)

Experimental and computed laminar burning velocities for the stoichiometric CH_4-N_2O mixture diluted with 50% inert at p_0 = 1.0 bar and T_0 = 300 K:

Inert	Su,exp (cm/s)	Su,calc (cm/s)
He	70	53
Ar	48	34
N_2	32 [4]	20 [4]
CO_2	11	14*

* calculated value for mixture diluted with 40% CO_2.

Comparison of maximum flame temperature, heat release rate, and peak concentrations of main reaction intermediates (*Xi*: H, O, OH, HO_2), of stoichiometric CH_4-N_2O mixtures diluted with 50% inert (He, Ar, and N_2) and 40% CO_2 at p_0 = 1.0 bar and T_0 = 300 K:

Inert	$T_{f,max}$ (K)	-dQ/dt*10⁹ (J s⁻¹ m⁻³)	X_H *10⁴	X_O *10³	X_{OH} *10³	X_{HO2} *10⁶
50% He	2628	8.66	2.342	1.640	9.44	16.16
50%Ar	2662	6.85	1.092	0.766	4.87	7.94
50% N_2	2450	3.48	0.731	0.529	3.79	6.98
40% CO_2	2257	2.29	0.335	0.498	3.28	7.91

Baric coefficients, -β, and overall reaction orders, n, at T_0 = 300 K, for the stoichiometric CH_4-N_2O mixture diluted with 50% inert:

Inert	β from Su,exp	Overall reaction order, n
He	-0.010	1.98
Ar	-0.066	1.89
N_2	-0.156 [4]	1.69 [4]
CO_2	-0.096	1.81

Baric coefficients, -β: $S_u = S_{u,ref}\left(\frac{p}{p_{ref}}\right)^{\beta}$

Overall reaction order, n, at T_0 = 300 K: $n = 2(\beta + 1)$

CONCLUSIONS

- The pressure influence on the laminar burning velocities of stoichiometric methane–nitrous oxide diluted with 50 vol% inert (He, Ar, N_2 or CO_2) was examined and the corresponding baric coefficients were obtained.
- The computed laminar burning velocities are lower when compared to experimental values.
- The overall reaction orders, n, at T_0 = 300 K range between 1.69 when nitrogen is used as inert, and 1.98 when helium is used as inert.
- The influence of additive type (He, Ar, N_2 or CO_2) on laminar burning velocities, maximum flame temperature, heat release rate and peak concentrations of main reaction intermediates was investigated and presented.
- Among the studied additives, CO_2 is the most efficient, followed by N_2, Ar, and He.
- The present results have a great practical interest for predicting the laminar burning velocity of stoichiometric methane–nitrous oxide at initial pressures at the ambient initial temperature, in the presence of various diluent gases.

References

1. Razus, D.; Oancea D.; Movileanu, C. *Journal of Loss Prevention in the Process Industries*, **2006**, *19*, 334-342.

2. Mitu, M.; Razus, D.; Giurcan, V.; Oancea, D. *Fuel,* **2015**, *147*, 27–34.

3. Cosilab, version 3.0.3., Rotexo-Softpredict-Cosilab GmbH & Co KG, Bad Zwischenhahn, **2011**.

4. Razus, D.; Mitu, M.; Giurcan, V.; Movileanu, C.; Oancea, D. *Process Safety and Environmental Protection,* **2018**, *114*, 240-250.