

## Article

# Propagation of CH<sub>4</sub>-N<sub>2</sub>O-N<sub>2</sub> Flames in a Closed Spherical Vessel

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**Abstract:** Flammable fuel-N<sub>2</sub>O mixtures raise safety and environmental protection issues in areas where these mixtures are used (such as: industry, research, internal combustion engines). Therefore, it is important to know their laminar combustion velocities and propagation speeds—important safety parameters for design of active protection devices against gas explosions and corresponding safety recommendations. In this paper, the laminar combustion velocities of N<sub>2</sub>-diluted CH<sub>4</sub>-N<sub>2</sub>O flames, obtained in experiments on outwardly propagating flames, at various initial pressures (within 0.5–2.0 bar) and room temperature, are reported. The experiments were made in a 0.5 L spherical cell with central ignition. The laminar combustion velocities were calculated from the constants of cubic law of flame propagation during the early stage of closed cell explosions and the expansion coefficients of unburned flammable mixtures, using the adiabatic model of the flame propagation. The expansion coefficients were determined from equilibrium calculations on flames propagating under isobaric conditions. The laminar combustion velocities were compared with data reported in the literature. Using the laminar combustion velocities and the expansion coefficients, the propagation speeds of N<sub>2</sub>-diluted CH<sub>4</sub>-N<sub>2</sub>O flames were calculated. Both laminar combustion velocities and propagation speeds decrease with the initial pressure increase.

**Keywords:** laminar combustion velocity; propagation speed; methane; nitrous oxide; closed cell combustion



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## 1. Introduction

Determination of flammability properties of gaseous combustible mixtures in various conditions is the key to achieving the performance of combustion systems and the safety in their use. Special attention is paid to flammability limits, maximum explosion pressures, explosion indices (severity factors) and laminar combustion velocities of fuel-air gaseous mixtures. Presently, studies on the reaction characteristics were conducted not only on fuel-air mixtures but also on fuel-nitrous oxide mixtures, motivated by the recent interest in using nitrous oxide as an oxidant in propulsion systems [1–3]. Nitrous oxide is an oxidizer with enhanced oxidizing potential compared with air, but its handling is as critical as is handling pure oxygen, since N<sub>2</sub>O is extremely exothermic and an additional heat amount is released during its decomposition. The nitrous oxide can behave as both as oxidizer and it can act as an explosive when it dissociates exothermically [4]. It is generated by waste incineration, radiolysis, or chemical reactions. N<sub>2</sub>O can be observed in nuclear waste storage facilities, together with methane or hydrogen and therefore they can form easily a flammable mixture. Thus, flammable fuel-N<sub>2</sub>O mixtures raise safety and environmental protection issues in areas where these mixtures are used.

The combustion of fuels in presence of the nitrous oxide can produce highly unstable and turbulent deflagrations that rapidly self-accelerate and could change from deflagration to detonation regime. Bane et al. [5] highlighted that nitrous oxide flames become unstable

immediately after ignition, even at low pressures and at lean compositions of flammable mixtures. Although there are many regulations to protect workers in the combustion field, there are accidental explosions related to fuel-N<sub>2</sub>O mixtures, some of them with disastrous consequences, due to the wider explosion limits and lower ignition energies of these mixtures. Therefore, such studies are needed to limit accidents. However, compared with fuel-air or fuel-oxygen mixtures, there are fewer studies on fuel-nitrous oxide reaction systems, especially on their laminar combustion velocities.

Several studies were conducted in the past to provide information on explosions in fuel-N<sub>2</sub>O mixtures. Some authors [6–15] reported the explosion parameters such as the flammability limits, the maximum explosion pressures, the explosion times, or the deflagration indices for explosions occurring in closed cells in the presence of N<sub>2</sub>O as oxidizer. The laminar combustion velocity of fuel-N<sub>2</sub>O and fuel-N<sub>2</sub>O-diluent mixtures (fuels: NH<sub>3</sub>, CO, H<sub>2</sub>, C<sub>2</sub>H<sub>4</sub> and C<sub>2</sub>H<sub>2</sub>) was investigated mainly by burner method [16–19]. In addition, other authors used the closed vessel method to obtain the laminar combustion velocities of H<sub>2</sub>-N<sub>2</sub>O [20–22], C<sub>2</sub>H<sub>4</sub>-N<sub>2</sub>O or C<sub>2</sub>H<sub>4</sub>-N<sub>2</sub>O-N<sub>2</sub> mixtures [23]. These results were helpful to provide data for testing and developing comprehensive chemical models of fuel-nitrous oxide combustion.

A widely examined fuel is methane, the main component of natural gas or mine gas. Methane is also the main flammable component of biogas, a low-cost, sustainable, renewable fuel produced by action of microorganisms on waste from vegetation, human and animal biological activity. Even if most previous research simply focused on the explosion of methane using air as oxidizer [24–30], the recent studies have also turned their attention to the explosive behavior of methane using N<sub>2</sub>O as oxidizer. Experiments using stationary flames (the burner method [31–33] or the counter-flow twin flames method [34]) or non-stationary flames (the constant volume method [35–37]) and numerical modeling were conducted using CH<sub>4</sub>-N<sub>2</sub>O gaseous mixtures (usually in the presence of diluents) to expand the available database regarding these flames.

Although the CH<sub>4</sub>-N<sub>2</sub>O-diluent flames were frequently studied, to date there is little data regarding their laminar combustion velocities and propagation speeds at pressures higher than ambient. In this research, the laminar combustion velocities of CH<sub>4</sub>-N<sub>2</sub>O-N<sub>2</sub> flames are calculated using the constants of cubic law of the flame propagation during the early stage of explosion and the severity coefficients of unburned flammable mixtures, using the adiabatic model of the flame propagation [38]. The motivation of the present research rises from the necessity to predict and reduce the risks associated with fires and explosions in industrial facilities and research laboratories that handle and use nitrous oxide, an oxidizer with enhanced oxidizing potential.

The method approached in the present study is useful to evaluate the laminar combustion velocity of mixtures containing a composite fuel (i.e., diesel, gasoline, landfill gas or liquefied petroleum gas – LPG) or composite additives (i.e., flue gases), from records of pressure-time evolution during explosions in an enclosures, avoiding calculations performed using equations that contain parameters unavailable for mixtures of complex composition, such as the adiabatic flame temperature and pressure [38]. Because this calculation method involves only calculable parameters (the adiabatic compression coefficient and severity coefficient) along with the cubic law coefficients, it is suitable for determining the laminar combustion velocity for mixtures at different initial conditions.

Using the laminar combustion velocities and the severity coefficients, the propagation speeds of N<sub>2</sub>-diluted CH<sub>4</sub>-N<sub>2</sub>O flames were calculated. For both laminar combustion velocities and propagation speeds the pressure dependence is examined and discussed.

The data obtained in this research could share new perspectives regarding the explosion phenomena when nitrous oxide is used as oxidizer since N<sub>2</sub>O is more energetic compared with oxygen or air. On the other hand, the present results help develop explosion protection measures for handling, storage and use of these mixtures in industrial and domestic installations.

## 2. Experimental Procedure

Experiments were conducted in a spherical enclosure with a radius of 5 cm ( $V = 0.5$  L) and central ignition, using stoichiometric and lean ( $\varphi = 0.8$ )  $\text{CH}_4\text{-N}_2\text{O-N}_2$  mixtures at different initial pressures between 0.5 and 2.0 bar and room temperature. In these small-scale explosions, the flame front of reactive mixtures does not develop significant cellular structures, a fact that supports the use of this calculation model. The studied  $\text{CH}_4/\text{N}_2\text{O}$  ratios, as shown by these equivalence ratios, were chosen as representative since they afford the complete fuel consumption and the comparison with other data sets from literature. For safety concerns, the  $\text{CH}_4\text{-N}_2\text{O}$  mixtures were diluted with various amounts of  $\text{N}_2$ : 40, 50 and 60 vol% respectively, expressed as percentages of the total mixture. The mixtures prepared at a total pressure of 4 bar in a 10 L metallic cylinder according to the partial pressure method were used 48 h after mixing.

A schematic diagram of the experimental installation used in this research is illustrated in Figure 1.

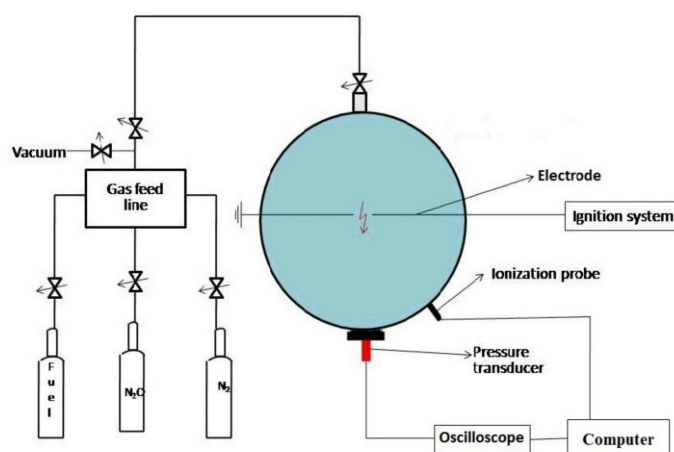


Figure 1. Schematic diagram of the experimental installation.

The experimental enclosure is equipped with a control unit, a measurement and control system, a pressure measuring system, a vacuum pump, and an ignition device. Before each explosion test, the experimental cell was cleaned and vacuumed down to 0.1 mbar. After the mixture was added to the vessel, sparks produced between two stainless steel electrodes ignited the mixture. The spark gap located in the geometrical center of the vessel had a constant width of 3.5 mm. After ignition, the pressure transient during explosions is recorded using a piezoelectric pressure transducer (Kistler 601A, Kistler, Winterthur, Switzerland), coupled with a Charge Amplifier (Kistler 5001SN, Kistler, Winterthur, Switzerland) and an Acquisition Data System Tektronix TestLab 2505. Minimum three experiments were performed at each investigated initial pressure, in each case with fresh mixture, to ensure the accuracy and repeatability of the data. The errors in the pressure measurement were up to 3.5%. Methane (99.99%),  $\text{N}_2\text{O}$  (99.999%) and nitrogen (99.99%), purchased from SIAD Italy, were used without further purification. Details of the experimental installation and procedure can also be found elsewhere [39–41].

## 3. Computing Program

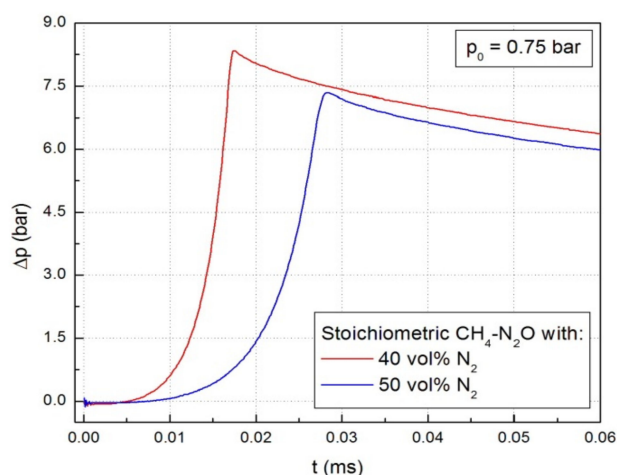
The characteristic parameters of isobaric (the burned (end) gas volume and flame temperature) and isochoric (the flame temperature and peak explosion pressure) adiabatic combustion were obtained using COSILAB 0-D package [42]. The package allows the computation of the equilibrium composition of products for any fuel-oxidizer gaseous mixture using the minimum of free Gibbs energy (at constant pressure) and the minimum of the free Helmholtz energy (at constant volume) as thermodynamic criteria of chemical equilibrium. Fifty-three compounds were considered combustion products. The thermodynamic data were given according to the international standard format, put forward by Sandia National

laboratories. The runs were performed for stoichiometric and lean ( $\varphi = 0.8$ )  $\text{CH}_4\text{-N}_2\text{O}$  flames in the presence of  $\text{N}_2$  at room temperature and various initial pressures (0.5–2.0 bar). Other details can be found in previous papers [43,44].

#### 4. Results and Discussion

The laminar combustion velocity is defined as the linear velocity of the flame front normal to itself relative to unburned gas, or as the volume of unburned gas consumed per unit time divided by the area of the flame front in which that unburned gas volume is consumed. Straightforward procedures are necessary to obtain the laminar combustion velocity of stationary premixed flames, but their use is limited to experiments at ambient pressure. The determination of the laminar combustion velocities at pressures above 1 bar is made using experiments produced in closed cells, using methods for synchronous recording the pressure and flame radius, along with adequate correction procedures meant to consider the flame stretch [22,23]. A simple procedure for determining laminar combustion velocities only from the pressure variation during the early stage of closed cell explosions was earlier developed using either models of an isothermal or adiabatic propagation [38]. Most data were obtained from the isothermal model, which delivered reliable results for numerous gaseous mixtures, under variable initial composition, pressure, and temperature [37–39,45–47]. An alternative way is to use the adiabatic model of flame propagation that gives correlates the coefficient of cubic law of pressure variation during the early stage of flame propagation and its expansion coefficient with the laminar combustion velocity [37,48]. In the present research, this way was used to calculate the laminar combustion velocities of  $\text{N}_2$ -diluted  $\text{CH}_4\text{-N}_2\text{O}$  mixtures and further on, their propagation speeds.

Typical diagrams of the pressure variation in the spherical vessel during the explosion of stoichiometric  $\text{CH}_4\text{-N}_2\text{O}$  mixture diluted with  $\text{N}_2$  (40% and 50%, respectively), at  $p_0 = 0.75$  bar and room temperature are illustrated in Figure 2. Similar graphs were obtained at all initial pressures.



**Figure 2.** Pressure variation during explosions in stoichiometric  $\text{CH}_4\text{-N}_2\text{O}$  mixture diluted with various  $\text{N}_2$  amounts, at  $p_0 = 0.75$  bar and room temperature.

Restricting the examination of the flame propagation only to the early stage, when the temperature gradients in both unburned and burned gas are small and can be neglected and when the flame maintains its spherical shape and the heat losses do not influence its development, a simple equation correlates the variation of pressure rise ( $\Delta p$ ) with time from ignition ( $t$ ) [38]:

$$\Delta p = a + k(t - b)^3 \quad (1)$$

where  $k$  is the cubic law constant;  $a$  and  $b$  are pressure and time corrections, respectively. These corrections are necessary to eliminate the errors in time and pressure measurements, respectively. Equation (1) is considered valid only for a restricted range of pressure

variation:  $\Delta p \leq p_0$  [38]. Considering an adiabatic compression of unburned gas ahead of the flame front, the laminar combustion velocity,  $S_u$ , is obtained using the following relationship [38]:

$$S_u = R \left[ \frac{k}{p_0 E^2 (E - 1) \gamma_u} \right]^{1/3} \quad (2)$$

here,  $R$  is the radius of the explosion enclosure;  $E$  is the expansion coefficient characteristic to the early (initial) stage of fuel-oxidizer explosion;  $p_0$  is the initial pressure and  $\gamma_u$  is the adiabatic compression coefficient of the unburned mixture.

It is worth mentioning that the present method requires the cubic law constants of pressure rise during the early stage of explosions, along with calculable properties such as the expansion coefficient and the adiabatic compression coefficient. This feature recommends this method for evaluation of early stage pressure variation in closed cells with various shapes and symmetry aspects, as used in many laboratory-scale explosion studies.

The coefficients  $k$  from Equation (1) were obtained for each experiment using a nonlinear regression method. Representative values of these coefficients, under various initial conditions, are given in Figure 3. They depend on many factors such as initial pressure, fuel, and diluent content. At constant initial temperature, fuel and dilution, the constants of cubic law increase with an increase in the initial pressure, as shown in Figure 3 for lean- and stoichiometric  $\text{CH}_4\text{-N}_2\text{O}$  mixtures diluted with  $\text{N}_2$ .

For any flammable mixture, the expansion coefficient defined as the ratio of burned and unburned gas volumes or the ratio of unburned to burned gas densities across the flame front can be obtained by equilibrium calculations:

$$E = \frac{V_e}{V_0} \quad (3)$$

where  $V_0$  is the volume of reactants and  $V_e$  is the volume of burned (“end”) gas generated by combustion of  $V_0$ .

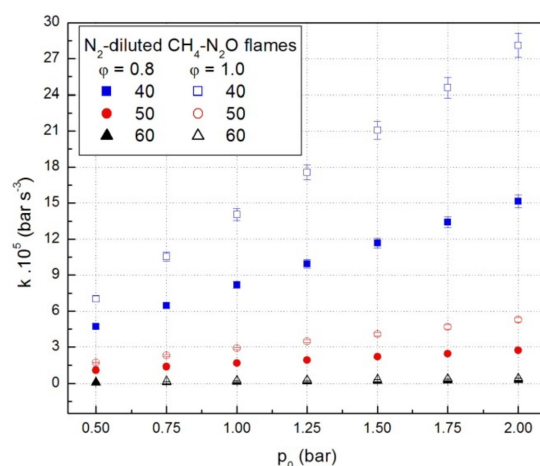


Figure 3. Cubic law constants of deflagrations in  $\text{CH}_4\text{-N}_2\text{O}$  mixtures diluted with  $\text{N}_2$ .

For  $\text{N}_2$ -diluted  $\text{CH}_4\text{-N}_2\text{O}$  mixtures the expansion coefficients delivered by equilibrium calculations on their adiabatic isobaric combustion are given in Table 1. At each composition, the expansion coefficients increase slightly with the initial pressure; at constant initial pressure, the expansion coefficients decrease with the increase of the diluent content.

**Table 1.** Expansion coefficients of deflagrations in N<sub>2</sub>-diluted CH<sub>4</sub>-N<sub>2</sub>O mixtures.

$p_0$ (bar)	$\varphi = 0.8$			$\varphi = 1.0$		
	40 vol% N <sub>2</sub>	50 vol% N <sub>2</sub>	60 vol% N <sub>2</sub>	40 vol% N <sub>2</sub>	50 vol% N <sub>2</sub>	60 vol% N <sub>2</sub>
0.50	10.87	9.80	8.54	11.21	10.17	8.97
0.75	10.97	9.85	8.56	11.29	10.23	9.01
1.00	10.99	9.88	8.57	11.35	10.27	9.03
1.25	11.03	9.91	8.58	11.40	10.31	9.05
1.50	11.05	9.92	8.58	11.44	10.33	9.06
1.75	11.08	9.94	8.59	11.47	10.36	9.08
2.00	11.10	9.95	8.59	11.49	10.42	9.09

Flamm and Mache [49], Lewis and von Elbe [50], Babkin et al. [51], and Agrawal [52] suggested alternative relationships for calculating the expansion coefficients, when no equilibrium calculations are possible (e.g., flammable gases with complex composition). These equations are summarized in Table 2. They deliver approximate values of expansion coefficients, based on a parameter characteristic to the isochoric combustion, i.e., the maximum (end) pressure  $p_e$ , reached in explosions at  $p_0$ . It was noted that constants  $A$  and  $B$  (from Babkin's equation) were found to be weakly dependent on the state of the mixture and its composition [51], so that Agrawal's general equation is just a more simplified form of Babkin's equation.

**Table 2.** Approximate relationships for calculating the expansion coefficients, by data referring to the isochoric combustion of flammable mixtures.

Flamm and Mache [49] and Lewis and von Elbe [50]	Babkin et al. [51]	Agrawal [52]
$E = 1 + \frac{1}{\gamma_u} \left( \frac{p_e}{p_0} - 1 \right)$	$E = A + B \frac{p_e}{p_0}$ , equation simplified later as $E = 0.85 \frac{p_e}{p_0}$	$E = 0.848 \frac{p_e}{p_0}$

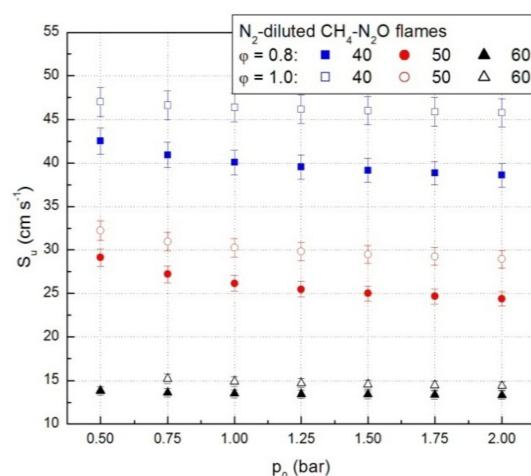
A comparison of results obtained using these equations is given in Table 3, for two sets of initial conditions. For all data sets, the adiabatic explosion pressures were used instead of experimental peak explosion pressures, dependent on real, non-adiabatic conditions of propagation. This affords a better comparison of data given in Table 3. In all examples shown in this table the expansion coefficients obtained by equations Babkin-Agrawal are close to those delivered by the exact computations, while the values computed with the approximate equation of Flamm-Mache-Lewis-von Elbe are lower for all compositions. This prompted us to use only the expansion coefficients calculated by Equation (3) for calculating the laminar combustion velocities, using the constants of cubic law and the adiabatic compression model.

**Table 3.** Expansion coefficients of CH<sub>4</sub>-N<sub>2</sub>O mixtures diluted with N<sub>2</sub>

$p_0$ (bar)	Lean ( $\varphi = 0.8$ ) CH <sub>4</sub> -N <sub>2</sub> O-60%N <sub>2</sub> Mixture (the Slowest Studied Mixture)			Stoichiometric CH <sub>4</sub> -N <sub>2</sub> O-40%N <sub>2</sub> Mixture (the Fastest Studied Mixture)		
	Equation (3)	Equation Flamm-Mache- Lewis-von Elbe	Equation Babkin-Agrawal	Equation (3)	Equation Flamm-Mache- Lewis-von Elbe	Equation Babkin-Agrawal
0.50	8.54	7.81	8.62	11.21	10.17	11.12
0.75	8.56	7.85	8.66	11.29	10.26	11.22
1.00	8.57	7.87	8.69	11.35	10.33	11.30
1.25	8.58	7.89	8.71	11.40	10.38	11.35
1.50	8.58	7.90	8.72	11.44	10.42	11.40
1.75	8.59	7.91	8.74	11.47	10.45	11.44
2.00	8.59	7.94	8.75	11.49	10.47	11.47



A set of representative results referring to lean  $\text{CH}_4\text{-N}_2\text{O}$  mixture diluted with  $\text{N}_2$  is plotted in Figure 4. Similar graphs were obtained for all  $\text{N}_2$ -diluted  $\text{CH}_4\text{-N}_2\text{O}$  mixtures.



**Figure 4.** Laminar combustion velocities of lean and stoichiometric  $\text{CH}_4\text{-N}_2\text{O-N}_2$  mixtures, from experimental  $p(t)$  data.

For all examined  $\text{N}_2$ -diluted  $\text{CH}_4\text{-N}_2\text{O}$  mixtures the laminar combustion velocities decrease with initial pressure, within the examined range of initial conditions. A decrease of the laminar combustion velocity at initial pressure increase was already reported in the literature for other flammable gaseous mixtures, under various initial conditions [25,37,39–41,43,44,53].

The increase in the initial pressure usually decreases the radical concentration within the reaction zone, along with the increase of the flame temperature. These decreases in the radical concentrations are responsible in a large extent for the reduction in the laminar combustion velocities with initial pressure increase. Numerous computational studies on hydrocarbons burning pointed out that the main chain carriers are the radicals  $\text{O}$ ,  $\text{H}$  and  $\text{OH}$ , which seem to most influence the fuel consumption reaction [37,40,45,54]. Another study [55] showed that  $\text{CO}$  and  $\text{NO}$  are also important for flame propagation in fuel- $\text{N}_2\text{O}$  mixtures.

The dependence of laminar combustion velocities on the initial pressure was analyzed using an empirical power law [38,43–47,56]:

$$S_u = S_{u,ref} \left( \frac{p}{p_{ref}} \right)^{\nu} \quad (4)$$

where  $S_{u,ref}$  is the laminar combustion velocity at reference pressure ( $p_{ref}$ ) and  $\nu$  is the baric coefficient. Choosing the atmospheric pressure as reference,  $p_{ref} = 1$  bar, the corresponding baric coefficients of laminar combustion velocities of the studied mixtures were obtained using a non-linear regression analysis of  $S_u = f(p)$ . The results are presented in Table 4, where the baric coefficients of laminar combustion velocities delivered by the kinetic modeling of  $\text{CH}_4\text{-N}_2\text{O-N}_2$  flames by using GRI 3.0 mechanism [37] are also given.

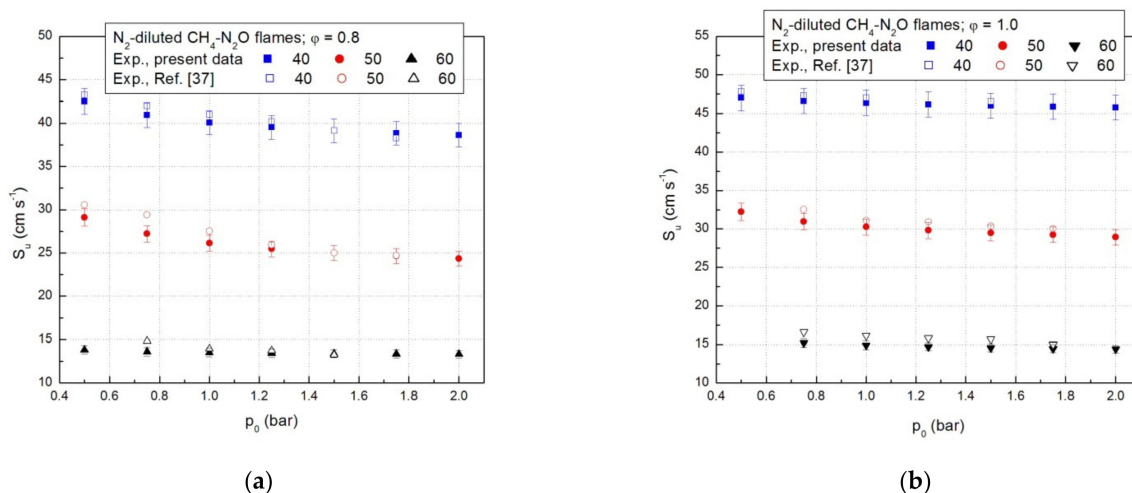
**Table 4.** The baric coefficients of laminar combustion velocities of the studied mixtures.

$\phi$	[N <sub>2</sub> ] (vol%)	-v	
		From Experimental $S_u$ (Present Data)	From Computed $S_u$ [37]
0.8	40	$0.072 \pm 0.004$	$0.097 \pm 0.010$
	50	$0.134 \pm 0.007$	$0.082 \pm 0.009$
	60	$0.027 \pm 0.002$	- *
1.0	40	$0.019 \pm 0.001$	$0.101 \pm 0.006$
	50	$0.078 \pm 0.005$	$0.119 \pm 0.009$
	60	$0.058 \pm 0.003$	$0.118 \pm 0.008$

\* no runs were possible at this composition.

Excepting the lean CH<sub>4</sub>-N<sub>2</sub>O mixture diluted with 50% N<sub>2</sub>, a low pressure dependence of the laminar combustion velocities is observed. This behavior was already observed by Bane et al. [5] for undiluted H<sub>2</sub>-N<sub>2</sub>O flames at pressures between 1 and 3 bar. They found global reaction orders close to 2 for these mixtures, a fact that explains the observed low pressure dependency of the laminar combustion velocities. Furthermore, the authors noticed that for a given N<sub>2</sub> dilution, there is no significant variation of the combustion speeds with the increase in the initial pressure. In addition, Movileanu et al. [46] reported a weak dependence of the laminar combustion velocity on the initial pressure for N<sub>2</sub>-diluted ethylene-air mixtures. The laminar combustion velocities reported in the present research can be compared with those reported in the literature.

In a previous study [37], the laminar combustion velocities of CH<sub>4</sub>-N<sub>2</sub>O-N<sub>2</sub> flames were calculated according to the model of isothermal flame propagation. A comparison between the present data and the laminar combustion velocities delivered by the isothermal model is given in Figure 5a,b. Within experimental errors, the laminar combustion velocities obtained from the adiabatic model of the flame propagation are close to those obtained by the isothermal compression model.



**Figure 5.** Laminar combustion velocities of CH<sub>4</sub>-N<sub>2</sub>O mixtures diluted with N<sub>2</sub>; present results and literature data: (a) for equivalence ratio  $\phi = 0.8$  and (b) for equivalence ratio  $\phi = 1.0$ .

Powell et al. [31–33] examined by experiments and modeling the laminar combustion velocities of stoichiometric CH<sub>4</sub>-N<sub>2</sub>O mixture in the presence of 52% N<sub>2</sub> at near atmospheric pressure (0.8 bar). They reported  $S_u = 24.2 \text{ cm s}^{-1}$  from measurements using a flat flame burner and  $S_u = 21.5 \text{ cm s}^{-1}$  from modeling using a modified PPD mechanism. In comparison, we found a higher value:  $S_u = 31.0 \text{ cm s}^{-1}$  for the stoichiometric CH<sub>4</sub>-N<sub>2</sub>O mixture with 50% N<sub>2</sub> at 0.75 bar (from experimental data).



Another study [57] was conducted on stoichiometric CH<sub>4</sub>-N<sub>2</sub>O mixture diluted with 40 and 50% N<sub>2</sub> using a Bunsen burner. The results are given in Table 5 together with the present data referring to the same mixtures at ambient initial pressure; here, the two sets of data agree well.

**Table 5.** Laminar combustion velocities of stoichiometric N<sub>2</sub>-diluted CH<sub>4</sub>-N<sub>2</sub>O mixtures.

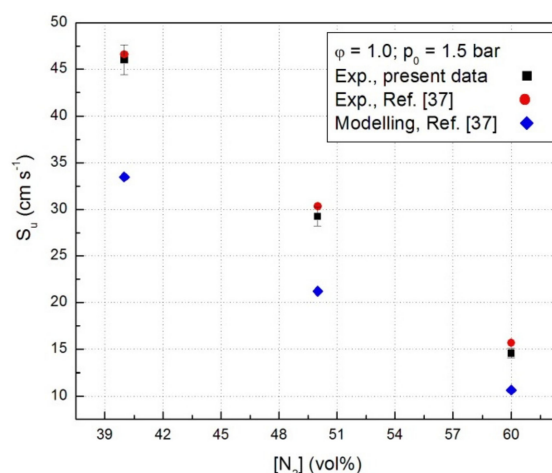
$S_u$ (cm s <sup>-1</sup> )		Reference
40% N <sub>2</sub>	50% N <sub>2</sub>	
46.4	30.3	Present data
46.5	30.1	[57]

For all systems, the computed laminar combustion velocities of CH<sub>4</sub>-N<sub>2</sub>O-N<sub>2</sub> flames were lower than the experimental  $S_u$  as shown in Figure 6 where data referring to stoichiometric CH<sub>4</sub>-N<sub>2</sub>O mixture in the presence of N<sub>2</sub> at constant initial pressure ( $p_0 = 1.5$  bar) are given. This was already reported for the laminar combustion velocities obtained for these mixtures from closed vessel experiments [37] or from flat flames, observed at 0.8 bar [32,33]. This trend was found for C<sub>2</sub>H<sub>4</sub>-N<sub>2</sub>O-N<sub>2</sub> mixtures as well, when the kinetic modeling of these flames has been made with GRI 3.0 mechanism, but no discrepancies were observed using other mechanisms with improved rate constants [23]. The sensitivity analyses undertaken by Powell et al. [32,33] revealed that several reactions within the hydrogen-N<sub>2</sub>O sub-mechanism are most important with respect to the laminar combustion velocity:



The improvement of their rate constants could lead to better predictions of the laminar combustion velocities.

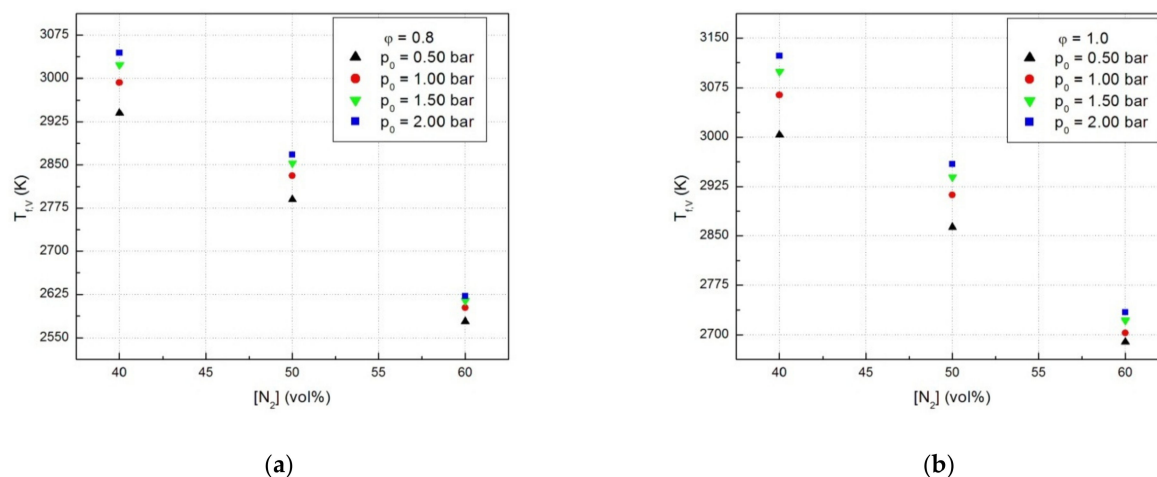
The presence of diluents in the flammable mixtures strongly affects both the kinetics and thermodynamics of the combustion process and consequently the laminar combustion velocity. As expected, the increase of the diluent content leads to the decrease of the laminar combustion velocity as it can be observed from Figure 6. The same trend was obtained at all initial pressures.



**Figure 6.** Laminar combustion velocities of stoichiometric N<sub>2</sub>-diluted CH<sub>4</sub>-N<sub>2</sub>O flames at constant initial pressure.

The decrease of the laminar combustion velocities determined by dilution with an inert gas can be linked with the decrease of the adiabatic flame temperatures of examined

mixtures. The plots of adiabatic flame temperatures ( $T_{f,v}$ ) in the isochoric combustion of  $N_2$ -diluted  $CH_4$ - $N_2O$  flames of constant equivalence ratio against the diluent content, shown in Figure 7a,b, have the same variation trend as the laminar combustion velocities, as already pointed out earlier [45].



**Figure 7.** Adiabatic flames temperatures of  $CH_4$ - $N_2O$  mixtures in the presence of  $N_2$ : (a) for equivalence ratio  $\phi = 0.8$  and (b) for equivalence ratio  $\phi = 1.0$ .

At constant initial pressure and temperature, adding nitrogen to a flammable mixture influences flame propagation through concentration changes (diminution of  $CH_4$  and  $N_2O$  concentrations), and through changes of reactivity. Dilution leads to diminution of available heat which sustain the explosion and hence, in the decrease of the temperature of the flame. Simultaneously,  $N_2$  addition significantly reduces the reaction rate, the rate of heat release and the total amount of radical species. Furthermore, in  $N_2O$  atmosphere, nitrogen is not acting just as an inert component. As outlined by Shen et al. [12],  $N_2$  can participate to consume O, inhibiting thus the decomposition of  $N_2O$  by by reaction (R.4):



An additional influence is exerted by  $N_2$  through its third-body collisional effect since it favors reaction (R.6) over the branching reaction (R.5), not mentioning the fact that  $HO_2$  is less reactive than O or OH [12]:



The laminar combustion velocities obtained in the present research together with the expansion coefficients can be used to calculate the propagation speeds,  $S_s$ , using the following equation:

$$S_s = S_{ll} \cdot E_0 \quad (5)$$

Relevant values of the propagation speeds obtained using Equation (5) are given in Figure 8. The pressure increase determines the decrease of propagation speeds. Simultaneously, the addition of a diluent also determines the decrease of the propagation velocities of the  $CH_4$ - $N_2O$ - $N_2$  mixtures by the decrease of the initial fuel concentration.

The propagation speed depends on the same parameters as the laminar combustion velocity (initial pressure and temperature, fuel type, fuel-oxidizer ratio, presence of diluents) [58,59] and, additionally, on the flow pattern. A power law characteristic for

flammable mixtures describes the dependence of the propagation speeds on the initial pressure [47]:

$$S_s = S_{s,ref} \left( \frac{p}{p_{ref}} \right)^a \quad (6)$$

In Equation (6)  $S_{s,ref}$  is the propagation speed at reference conditions (here  $p_{ref} = 1$  bar) and  $a$  is the baric coefficient. The baric coefficients of the propagation speeds, determined by a non-linear regression analysis of  $S_s = f(p)$  data, are presented in Table 6.

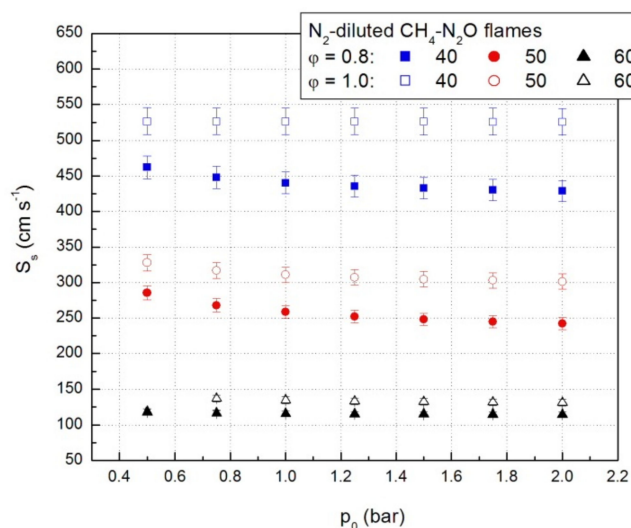


Figure 8. Propagation speeds of lean and stoichiometric CH<sub>4</sub>-N<sub>2</sub>O-N<sub>2</sub> mixtures.

Table 6. Baric coefficients of the propagation speeds of CH<sub>4</sub>-N<sub>2</sub>O-N<sub>2</sub> mixtures.

$\varphi$	[N <sub>2</sub> ] (vol%)	$-a$
0.8	40	$0.054 \pm 0.004$
	50	$0.118 \pm 0.007$
	60	$0.021 \pm 0.002$
1.0	40	$0.001 \pm 0.00004$
	50	$0.060 \pm 0.004$
	60	$0.047 \pm 0.003$

As shown by data from Table 6, a low pressure dependence of the propagation speed is observed, as in the case of laminar combustion velocities. The baric coefficients of propagation speeds do not differ much from those of laminar combustion velocities, since the pressure has a reduced influence on expansion coefficients.

The flammable mixtures of CH<sub>4</sub> and N<sub>2</sub>O raise much interest, as both components are present in the complex gaseous mixtures generated by nuclear wastes in the dome space of storage tanks. Moreover, the laminar combustion velocity and propagation speed, which are fundamental properties of any combustible mixture, are essential for the design of venting devices, combustors, engines (e.g., internal combustion or rocket engines), power plant burners etc. The laminar combustion velocity is also used in theoretical modeling of the pressure-time evolution inside a cell or a piping, which is important to consider during the design process.

## 5. Conclusions

The laminar combustion velocity and propagation speed are the most useful parameters for various applications important in industry and for safety, with impact on environmental protection. Simultaneously, the laminar combustion velocity and propagation speed are used to validate chemical reaction mechanisms and modeling the turbulent

combustion. Therefore, the accurate determination of the laminar combustion velocity and propagation speed is of great interest.

The combustion burning velocities of lean and stoichiometric  $\text{CH}_4\text{-N}_2\text{O}$  mixtures in the presence of  $\text{N}_2$ , at various initial pressures and room temperature, were obtained from the constants of cubic law of flame propagation during the early stage of closed cell explosions and from the expansion factors. The laminar combustion velocities and expansion coefficients were later used to calculate the propagation speeds of the  $\text{N}_2$  diluted  $\text{CH}_4\text{-N}_2\text{O}$  flames.

For the studied systems, a decrease of the laminar combustion velocities and hence of the propagation speeds was observed when the initial pressures increased. The laminar combustion velocity and the propagation speed dependence on initial pressure were examined by empirical power laws. The baric coefficients of these velocities were calculated by non-linear regression analysis of  $S_u = f(p)$  and  $S_s = f(p)$ , respectively.

The laminar combustion velocities were compared with those obtained from the isothermal model of flame propagation and with those obtained from the kinetic modeling of  $\text{N}_2$ -diluted  $\text{CH}_4\text{-N}_2\text{O}$  flames. The results reveal that both the isothermal model and the adiabatic compression model deliver higher values of laminar combustion velocities than those computed with GRI 3.0 mechanism.

The increase of the diluent content leads to the decrease of the laminar combustion velocities and the propagation speeds for all studied initial pressures and mixtures composition. The decrease of the laminar combustion velocities determined by dilution with an inert gas can be linked with the decrease of the adiabatic flame temperatures of examined mixtures and hence by the diminution of available heat able to sustain the explosion as well as the rate of heat release.

As the present method of laminar combustion velocity calculation needs only calculable quantities (expansion coefficient and adiabatic compression coefficient) along with the cubic law coefficients, this method is suitable for the evaluation of data recorded in cylindrical enclosures (centrally ignited) frequently used in industry and laboratory research.

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