



*Dedicated to Professor Ionel Haiduc
on the occasion of his 80th anniversary*

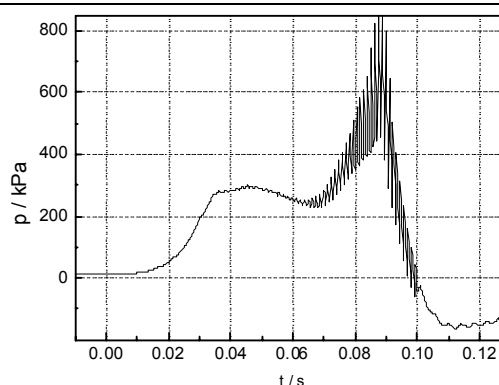
PROPAGATION INDEXES OF C₂H₄-N₂O-N₂ DEFLAGRATIONS IN ELONGATED CLOSED VESSELS

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Maximum explosion pressures, p_{max} , explosion times, θ_{max} and maximum rates of pressure rise, $(dp/dt)_{max}$, for lean and stoichiometric ethylene-nitrous oxide mixtures diluted with 60% N₂, at initial pressures p_0 within 50 - 150 kPa, measured in 3 cylindrical vessels with different aspect ratio, L/D (length to diameter) = 1.0; 1.5 and 8.1 are reported. For each closed vessel and each flammable composition, linear correlations of p_{max} and $(dp/dt)_{max}$ versus p_0 were found. The amount of heat transferred by the burned gas to the vessel, before the end of combustion, was determined for each flammable composition from the intercepts of p_{max} vs p_0 correlations.



INTRODUCTION

Nitrous oxide, N₂O, a less studied oxidizer, is used lately in rocketry and in motor racing to increase the power output of engines. Nitrous oxide is frequently used in surgery and dentistry for its anesthetic and analgesic effects. Despite these practical interests, there are few studies on fuel-nitrous oxide reaction systems as compared to fuel-air or fuel-oxygen. Early studies on N₂O flames reported the flammability range of various fuel-N₂O mixtures (the lower and upper flammability limits) in the presence of inert gases.¹⁻⁴ Combustion of hydrogen, carbon oxide, ammonia and other fuels (n-pentane, diethyl ether, diethylamine, n-butyraldehyde or toluene) with nitrous oxide has been investigated

for determining the main flammability characteristics of these mixtures (peak explosion pressures, severity factors, burning velocities) and the structure of these flames, necessary for understanding the combustion of nitrogen-containing energetic materials.⁵⁻⁹ The structure of N₂O flames with simple fuels (NH₃, CO, H₂, CH₄, C₂H₆, C₂H₂ or C₃H₈), burning in various conditions, has been reported^{6-8,10} together with their burning velocities. Measurements of temperature profiles and chemical species concentrations within the flame front (the main reaction zone) of fuel-N₂O flames stabilized in various burner configurations have been used to develop and test detailed kinetic models for fuel-nitrous oxide systems and other combustion systems involving nitrogen oxidation.

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Table 1

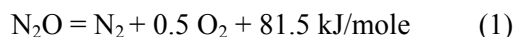
Characteristic properties of stoichiometric C₂H₄-air and C₂H₄-N₂O flames at $p_0 = 1$ bar and $T_0 = 300$ K, calculated from chemical equilibrium and kinetic modeling

Flame property	C ₂ H ₄ -air	C ₂ H ₄ -N ₂ O
$T_{f,p}$ / K	2369	3007
$T_{f,v}$ / K	2720	3568
$p_{max,ad}$ / MPa	0.934	1.941
S_u / (cm s ⁻¹)	83.4	102.0

Among simple fuels burning with N₂O, ethylene was less studied. Parker and Wolfhard¹¹ measured the burning velocity of C₂H₄-N₂O mixtures as a function of pressure in the range of atmospheric and sub-atmospheric pressures. Other works were focused on soot formation and growth in rich C₂H₄-N₂O laminar premixed flames at atmospheric pressure.^{12,13} DDT (Deflagration-to-Detonation) and detonation processes of C₂H₄-N₂O mixtures have been studied in a closed cylindrical vessel at initial pressures of 8-23 bar.⁹

A comparison of characteristic deflagration properties of C₂H₄-N₂O and C₂H₄-air with the same equivalence ratio revealed that N₂O flames develop much higher temperatures and higher explosion pressures in comparison with air flames. A set of relevant data on premixed flames of C₂H₄ with air or with N₂O at ambient initial conditions is given in Table 1, where the computed flame temperatures ($T_{f,p}$, the adiabatic flame temperature of isobaric combustion, and $T_{f,v}$, the adiabatic flame temperature of isochoric combustion) and adiabatic explosion pressures, $p_{max,ad}$, are listed together with normal burning velocities S_u obtained by chemical modeling.¹⁴

These features can be qualitatively explained by taking into account the reaction:



which releases an additional heat amount in fuel-N₂O flames in comparison to fuel-air flames.

For safety concern (to avoid detonation), our experiments were conducted in nitrogen-diluted C₂H₄-N₂O mixtures. The present contribution reports flammability properties (peak explosion pressure, maximum rate of pressure rise and time

to peak pressure) of nitrogen-diluted C₂H₄-N₂O mixtures in experiments performed at various initial pressures ($p_0 = 50$ -150 kPa), in 3 cylindrical vessels with L/D (length to diameter) = 1.0, 1.5 and 8.1. The flammability properties of C₂H₄-N₂O-N₂ mixtures are compared with those of C₂H₄-air mixtures having either the same ethylene concentration or an identical O/N ratio, to emphasize the large differences observed between the two systems.

EXPERIMENTAL

The experiments were performed in three cylindrical vessels with various length/diameter ratios; their geometric characteristics are listed in Table 2. The vessels were made from stainless steel and were equipped with ports for the gas feed/evacuation valve and for the ionization probes, pressure transducers and ignition electrodes. Each vessel was designed to withstand static pressures of at least 4 MPa. The ignition was made by high voltage inductive-capacitive sparks. In vessels C1 and C2 the spark gap was located in central position; in vessels C2 and C3 the spark gap was alternatively positioned in a point situated on the central axis, 15 mm above vessel bottom. The pressure variation was recorded with piezoelectric pressure transducers (Kistler 601A), in line with Charge Amplifiers Kistler 5001 SN. The signals from the ionization probe and from the pressure transducers were acquired at 2500 signals/channel and a maximum of 1 GS/s sampling rate, with a Tektronix digital oscilloscope type TDS 2014B connected to a PC. Other details on the experimental set-up and procedure have been previously described.¹⁵⁻¹⁷

Before each test, the combustion vessel was evacuated down to 0.5 mbar, the explosive mixture was admitted and allowed 10 min. to become quiescent, it was ignited and afterwards, evacuated. The gaseous mixtures were obtained by the partial pressure method and used 24 h after mixing the components, at a total pressure of 0.4 MPa.

Table 2

Geometric characteristics of explosion vessels

Name	Length / cm	Diameter / cm	Volume / cm ³	L/D
C1	6.0	6.0	170	1.0
C2	15.0	10.0	1180	1.5
C3	54.7	6.8	1986	8.1

Ethylene, purity 99.99%, N₂O and N₂, both of purity 99.999% (SIAD - Italy), have been used without further purification. Ethylene-N₂O mixtures with equivalence ratios ϕ^* between 0.8 and 1.0, diluted by 60% N₂ were used. Experiments have been made at initial pressures between 30 and 150 kPa and ambient initial temperature. Minimum 3 experiments have been performed for each initial condition of explosive mixture.

Computing programs and data evaluation

The adiabatic explosion pressures and the adiabatic flame temperatures have been calculated with the 0-D COSILAB package.¹⁸ The program is based on a general algorithm meant to compute the equilibrium composition of products for any fuel-oxidizer gaseous mixture using the thermodynamic criterion of chemical equilibrium: the minimum of Gibbs energy, at constant temperature and pressure or minimum of Helmholtz energy, at constant temperature and volume. Fifty-three compounds have been considered as combustion products.

The kinetic modeling of one-dimensional, premixed laminar free flames was performed with the COSILAB 1-D package (version 3.0.3) using GRI (Gas Research Institute) mechanism version 3.0 (53 chemical species and 325 elementary reactions).¹⁸ The input data were taken from thermodynamic and molecular databases of Sandia National Laboratories, USA, according to the international standard (format for CHEMKIN). The computations were performed for stoichiometric ethylene-air and ethylene-N₂O mixtures at ambient initial pressure and temperature.

The computation of (dp/dt) from $p(t)$ diagrams was made after smoothing the $p(t)$ data by Savitzky-Golay method, based on least squares quartic polynomial fitting across a moving window within the data. The method has the advantage of producing a smoothed first derivative without filtering the data. This involved the analysis of 500–700 points within ignition and the time necessary to reach the peak explosion pressure. In all cases, we used a 10% smoothing level, as higher values of this level (e.g. 20%) lead to a reduction of the noise accompanied by the signal distortion.

RESULTS AND DISCUSSION

Representative plots of pressure history for explosions of C₂H₄-N₂O-N₂ mixtures propagating in cylindrical vessels C2 and C3 are given in Figures 1 and 2.

In Fig. 1 a set of two $p(t)$ diagrams recorded in vessel C2 at central and bottom ignition are given. In both diagrams, a first stage of undisturbed flame propagation is observed, characterized by the highest rate of pressure rise of the whole process.

In the second stage of flame propagation, after the moment the flame has reached the side wall, a lower rate of pressure rise is observed, especially in experiments with bottom ignition. In Fig. 2 where data referring to the stoichiometric mixture are plotted, the early stage of fast pressure rise is followed by a pressure decrease and by a second peak of pressure where important pressure oscillations are recorded. As shown by Markstein¹⁹ and Phylaktou,²⁰ these three stages are characteristic for the flame propagation in elongated closed vessels. The first stage has usually a short duration (5-10% of the total explosion time) and it is characterized by the highest rate of pressure rise of the process. This high rate of pressure rise is related to the highest mean flame speed, characteristic of deflagrations in pipes. In the second stage, the pressure increase is almost linear and the flame speed is quasi-constant, depending on the equivalence ratio of the flammable mixture and on vessel geometry. During this stage, the cellular structure of flame is usually observed, revealing the onset of flame instability. In the last stage, the propagation speed is decreasing again and the flame instability increases. In these last two stages, the decrease of the maximum pressure, observed in experiments with bottom ignition, is due to a longer time of the heat transfer between hot combustion products and vessel walls.

For each flammable composition, the maximum explosion pressure was influenced by the initial pressure, by the dimension of the explosion vessel and by the position of the ignition source. Typical graphs p_{max} versus p_0 for centrally- and bottom-ignited explosions are shown in Figures 3 and 4, where the adiabatic explosion pressures were also plotted; for all graphs, the best-fit lines were drawn. In the case of bottom-ignited explosions in vessel C3 with $L/D = 8.1$ strong pressure oscillations have been observed for all examined C₂H₄-N₂O-N₂ mixtures. Average explosion pressures were measured in these cases, using the smoothed $p(t)$ diagrams. Data from Fig. 3 (centrally-ignited explosions) show that small differences are observed between the peak explosion pressures reached in vessels C1 ($L/D = 1.0$) and C2 ($L/D = 1.5$). Both data sets are inferior to the adiabatic explosion pressures, determined by unavoidable heat losses from the burned gases to the vessel. Similar plots are found in Fig. 4, where

* The equivalence ratio ϕ of C₂H₄-N₂O mixtures is defined as $\phi = \frac{([C_2H_4]/[N_2O])}{([C_2H_4]/[N_2O])_{stoich}}$ where the index "stoich" refers to the stoichiometric composition.

the peak explosion pressures reached in vessels C2 and C3 with bottom ignition are shown. As seen from Figures 3 and 4, linear correlations were found between the maximum explosion pressure and the initial pressure. Such correlations between the peak (maximum) explosion pressures and the initial pressures of fuel-air gaseous mixtures were reported for many explosions of quiescent mixtures, in deflagration regime, *e.g.* propane-air,¹⁶ LPG-air¹⁵ and propylene-air²¹ in a 0.5 L spherical vessel and in a 1.12 L cylindrical vessel.

$$p_{\max} = \left(\xi + \frac{\eta_l}{v_l} \cdot \frac{\Delta^c U'}{T_0 \cdot C_{e,V}} \right) \cdot p_0 - q_{tr} \frac{\gamma_e - 1}{V_0} = \pi_{ad} \cdot p_0 - q_{tr} \cdot \frac{\gamma_e - 1}{V_0} \quad (2)$$

where π_{ad} is the adiabatic coefficient of the pressure increase during explosion, defined as $\pi_{ad} = p_{ad,\max}/p_0$ and q_{tr} is the total amount of heat losses, transferred by the gas to the vessel before the end of combustion. The other terms are: $\xi = n_e/n_0$, the ratio of final to initial mole numbers; $\eta_l = n_l/n_0$, the ratio of the number of moles corresponding to the limiting component of the mixture and the total initial number of moles; v_l – the stoichiometric coefficient of the limiting component in the mixture; $\gamma_e = \overline{C_{e,p}}/\overline{C_{e,V}}$ – the adiabatic coefficient of the burned gas; $\overline{C_{e,p}}$ and $\overline{C_{e,V}}$ are the molar heat capacities (at constant p and constant V , respectively) of the end gaseous mixture, averaged for the end components and for the temperature range T_0 to $\overline{T_{e,V}}$; $\Delta^c U'$ is the molar combustion heat (at constant volume and T_0) corrected by

These data, as well as the present results, show a common feature: both the slope and intercept of the linear correlations are dependent on the initial composition of the flammable mixture and can be used to extract additional information on closed vessel combustion.

The examination of the heat balance during the constant volume combustion of a gaseous fuel-air mixture^{16,22} delivered a correlation of p_{\max} , the maximum explosion pressure, and the initial pressure p_0 :

taking into account the endothermic processes. It should be noted that the predictive power of equation (2) is limited by the implicit assumption that p_{\max} , the maximum explosion pressure, is equal to p_e , the pressure at the end of combustion, as previously shown.²²

Within a restricted pressure range, it was also assumed that ξ , $\Delta^c U'$ and $\overline{C_{e,V}}$ are independent of p_0 (or their variations compensate each other). According to this model, the amount of heat transferred by the burned gas to the vessel, before the end of combustion, for each flammable composition, can be determined from the intercepts of p_{\max} vs p_0 linear correlations. Comparative results concerning C_2H_4 - N_2O - N_2 and C_2H_4 -air flames are given in Table 3.

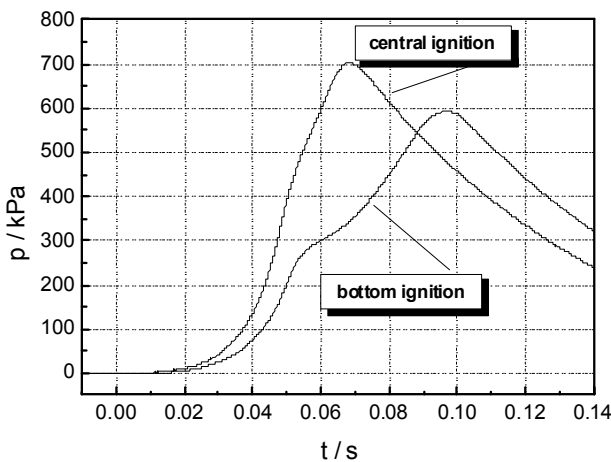


Fig. 1 – Pressure-time records for a lean C_2H_4 - N_2O - N_2 mixture ($\varphi = 0.80$) in vessel C2 ($L/D = 1.5$) with central or bottom ignition.

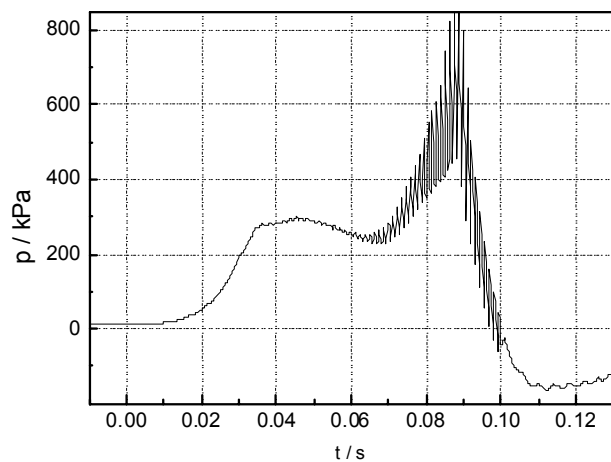


Fig. 2 – Pressure-time records for the stoichiometric C_2H_4 - N_2O - N_2 mixture in vessel C3 ($L/D = 8.1$) with bottom ignition.

It is observed that the heat losses appearing before the end of combustion are determined not only by the geometrical characteristics of the vessel and by the position of the spark gap, but also by the initial composition of the flammable mixture: higher heat amounts are lost to vessel walls by C₂H₄-N₂O flames, which are more exothermal in comparison with C₂H₄-air flames. The transferred heat amounts by the lean and stoichiometric C₂H₄-N₂O-N₂ mixtures ($\phi = 0.8$ and $\phi = 1.0$, respectively) during explosions in vessel C2 (175 and 342 J, respectively) reveal also the higher heat amount delivered by the stoichiometric mixture in comparison with the lean one.

The time to peak explosion pressure, θ_{max} , depends on the initial composition of the flammable mixture, on the position of the ignition source and on the volume and aspect ratio of the explosion vessel. Over the range of examined pressures, θ_{max} is less influenced by the initial pressure. Representative results on C₂H₄-N₂O-N₂ explosions in vessels C1-C3 are given in Table 4.

In any vessel, the stoichiometric C₂H₄-N₂O-N₂ mixture is characterized by shorter θ_{max} in comparison with the lean mixture, according to the burning velocities of these two mixtures.

For both centrally and bottom-ignited explosions, higher rates of pressure rise were observed in the early stage of explosion propagation (when a quasi-spherical flame without significant heat losses develops) as compared to later stages of the process, characterized by lower rates of pressure rise and influenced by the heat losses to the walls. Following the suggestion of Phylaktou,¹⁹ we name the rate of pressure rise measured in the early stage of propagation as “first rate of pressure rise” and the rate of pressure rise measured in the late stage of propagation as “second rate of pressure rise”. Representative results on the first rate of pressure rise for the examined flammable mixtures are shown in Figures 5 and 6. The maximum values of the first rate of pressure rise are indeed the highest rates of pressure rise for the whole process, relevant for adequate safety norms and for vent design. For each closed vessel and each flammable composition, linear correlations were found for the maximum rates of pressure rise against the initial pressure. As expected, the pressure rise rates of bottom-ignited explosions are always inferior to the rates of pressure rise rates for centrally ignited explosions.

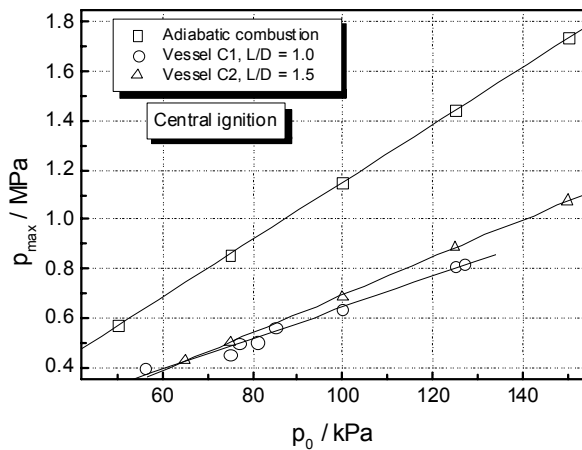


Fig. 3 – Peak explosion pressures of the stoichiometric C₂H₄-N₂O mixture diluted with 60 vol% N₂; central ignition.

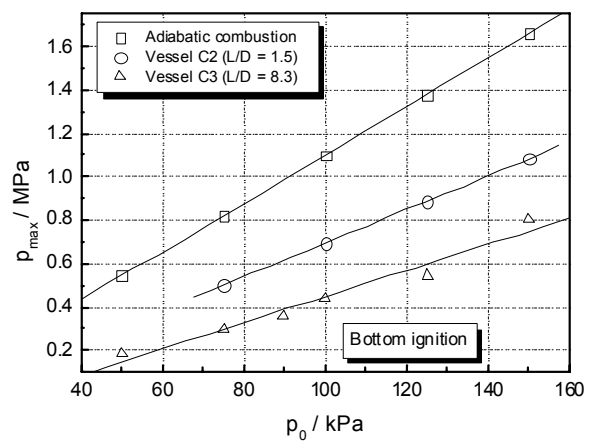


Fig. 4 – Peak explosion pressures of a lean C₂H₄-N₂O mixture ($\phi = 0.80$) diluted with 60 vol% N₂; bottom ignition.

Table 3

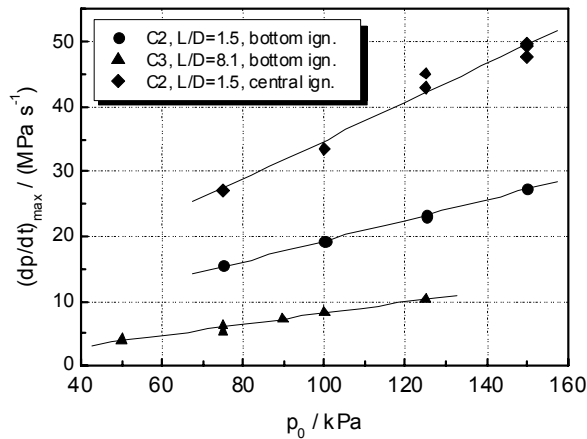
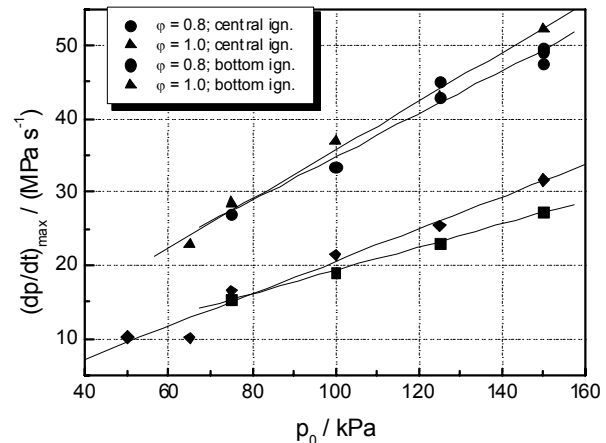
Heat lost, q_{tr} , by C₂H₄-N₂O-N₂ and C₂H₄-air flames to vessel walls, in centrally ignited explosions

Flammable mixture	q_{tr} / J	
	Vessel C1	Vessel C2
C ₂ H ₄ -N ₂ O-N ₂ with $\phi = 0.8$; [C ₂ H ₄] = 4.71 vol%	41.1	175
C ₂ H ₄ -air with [C ₂ H ₄] = 4.71 vol%	-	160
C ₂ H ₄ -N ₂ O-N ₂ with $\phi = 1.0$; [C ₂ H ₄] = 5.71 vol%	7.0	342
C ₂ H ₄ -air with [C ₂ H ₄] = 5.71 vol%	-	152

Table 4

Times to peak explosion pressure in experiments performed on $C_2H_4-N_2O-N_2$ mixtures at ambient initial conditions, θ_{max} / ms

Equivalence ratio, ϕ	Central ignition		Bottom ignition	
	Vessel C1	Vessel C2	Vessel C2	Vessel C3
0.8	35.1	68.2	96.2	152
1.0	29.4	50.2	69.3	88.6

Fig. 5 – Maximum rates of pressure rise for a lean $C_2H_4-N_2O-N_2$ mixture, $\phi = 0.80$; central or bottom ignition.Fig. 6 – Maximum rates of pressure rise in vessel C2 ($L/D = 1.5$); lean- and stoichiometric $C_2H_4-N_2O-N_2$ mixtures.

In experiments performed in vessel C3 ($L/D = 8.1$), strong pressure oscillations were observed in the later stage of explosions. Their amplitude depends on mixture strength and pressure and on the amount of added inert. The higher values of the pressure rise rate for the stoichiometric mixture in combination with these oscillations of the pressure signal indicate the strong flame acceleration along the tube. In less diluted mixtures, the flame acceleration can easily produce the transition to a detonation. Experiments made with undiluted $C_2H_4-N_2O$ mixtures in elongated vessels showed that a detonation could be initiated directly by using an energetic ignition source.⁹ By using a “mild”, low-energy source, only a deflagration was initiated as long as the pressure was under 800 kPa. Even so, the deflagration of undiluted $C_2H_4-N_2O$ mixtures underwent a fast acceleration and the DDT was observed.

The data update and complete the scarce information on deflagrations propagating in $C_2H_4-N_2O$ gaseous mixtures in elongated closed vessels where specific heat losses occur, strongly dependent on the vessel geometry and ignition point.

CONCLUSIONS

In the present study, characteristic parameters of confined explosions (p_{max} , θ_{max} and $(dp/dt)_{max}$) for 60% nitrogen-diluted stoichiometric $C_2H_4-N_2O$

mixtures are reported and discussed in correlation with the total initial pressure and composition of flammable mixtures, and with the aspect ratio of the explosion vessel and position of ignition source. The nitrogen-diluted $C_2H_4-N_2O$ mixtures have higher explosion pressures and rates of pressure rise and shorter times to peak explosion pressure in comparison to C_2H_4 -air mixtures with the same ethylene concentration. This is assigned to the exothermal dissociation of nitrous oxide in $C_2H_4-N_2O-N_2$ flames, releasing an additional heat amount in fuel- N_2O flames in comparison to fuel-air flames.

For all flammable mixtures, the measured peak explosion pressures are lower as compared to computed explosion pressures as a consequence of heat losses during the late stage of flame propagation. Higher heat losses are associated to a higher aspect ratio of the explosion vessel and to the asymmetrical position of the ignition source (earlier contact of flame with bottom and side walls, as compared to central ignition).

Linear correlations between the peak explosion pressure and the total initial pressure of flammable mixture were found for all examined mixtures. Both the slope and intercept of such correlations are influenced by the volume and aspect ratio of explosion vessel, and by the composition of the flammable mixtures. The intercept of $p_{max} = f(p_0)$ lines delivered the amount of heat lost to the walls

before the end of combustion. As expected, it is higher in C₂H₄-N₂O flames in comparison to C₂H₄-air flames with the same ethylene concentration and higher in stoichiometric C₂H₄-N₂O-N₂ flames in comparison to lean C₂H₄-N₂O-N₂ flames.

Linear correlations of the maximum rate of pressure rise against the total initial pressure were also found when the early stage of undisturbed flame propagation in each vessel was examined. The pressure rise rates of bottom-ignited explosions were always inferior to the rates of pressure rise rates for centrally ignited explosions.

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