Explosion pressures of hydrocarbon–air mixtures in closed vessels

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Abstract

An experimental study on pressure evolution during closed vessel explosions of several gaseous fuel–air mixtures was performed, at various initial pressures within 0.3–1.2 bar and ambient initial temperature. Explosion pressures and explosion times are reported for methane–, n-pentane–, n-hexane–, propene–, butene–, butadiene–, cyclohexane– and benzene–air mixtures. The explosion pressures measured in a spherical vessel (Φ = 10 cm) and in three cylindrical vessels with different diameter/height ratios are examined in comparison with the adiabatic explosion pressures, computed by assuming chemical equilibrium within the flame front. The influence of initial pressure, fuel concentration and heat losses during propagation (determined by the size and shape of the explosion vessel and by the position of the ignition source) on explosion pressures and explosion times are discussed for some of the examined systems.

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1. Introduction

Knowledge of pressure–time variation during explosions of fuel–air mixtures in enclosures is a very important component of safety recommendations for a wide range of human activities, connected to production, transportation or use of fuels.

The characteristic parameters of a closed vessel explosion are the explosion pressure, the explosion time and the maximum rate of pressure rise. The explosion pressure and explosion time were recently defined in the European standard on maximum explosion pressure determination [1]: (i) the explosion pressure, \( P_{\text{exp}} \) is the highest pressure reached during the explosion in a closed volume at a given fuel concentration; (ii) the maximum explosion pressure, \( P_{\text{max}} \) is the highest pressure reached during a series of explosions of mixtures with varying fuel concentration; (iii) the explosion time, \( \theta_{\text{exp}} \) is the time interval between ignition and the moment when the explosion pressure is attained.

Values of explosion pressures and explosion times of gaseous mixtures at various initial pressures and temperatures have been reported in many publications. Such values were obtained from measurements made in a wide set of conditions (explosion vessels with various forms and volumes; ignition made by local sources with various energies, from a few mJ up to 20 J; position of the ignition source: central or asymmetric) [2–23]. A great interest was also shown towards calculation of adiabatic explosion pressures reached in constant volume combustion, by using available programs for thermochemical equilibrium calculations [24–26] or by a detailed modeling of combustion coupled with heat and mass transfer processes [27–32].

Explosion pressures and explosion times are important also for calculating laminar burning velocities from closed vessel experiments [33–38], vent area design [8,17,27,31,39–41] and characterizing transmission of explosions between interconnected vessels [42–44].

Recent data on gas explosions were obtained from experiments in a spherical 20 L closed vessel with central ignition, produced by a fusing wire, a pyrotechnical ignitor or capacitive electric sparks. In fact, this is the vessel recommended by the recent European standards, for flammability [45] and explosion pressure [1] measurements. Few data measured according to European standard [1] are however available, especially for mixtures at pressures and/or temperatures different from ambient. Recent publications report data on methane–air mixtures both at ambient initial conditions and at elevated temperatures and pressures, on propene–oxygen and propene–air at normal and elevated pressures [19], on n-butane–oxygen at elevated...
pressure and temperature [23], on benzene–air, methanol–air and benzene/methanol–air mixtures at 150 °C and atmospheric initial pressure [22] from measurements in a 20 L apparatus. Values of maximum explosion pressures for gaseous hydrocarbons in the presence of air at ambient initial conditions, measured in the last few years by means of the recommended method [1], are given in the German collections of data [18,20,46]. These sources of data do not include, however, the influence of fuel concentration in fuel–air mixtures or the influence of oxidant/inert concentration in fuel–oxidant–inert mixtures on explosion pressures. Another recent publication reported measurements performed both in a 20 L vessel and in two larger vessels, of 120 L and 25.5 m³ volume [14]. The authors tried to evaluate the influence of heat losses during explosion development on explosion pressure and maximum rate of pressure rise for methane–, propane– and hydrogen–air mixtures. The maximum explosion pressures were lower in this vessel as compared to the standard one (V = 20 L), for all examined systems.

In the present paper, data on constant volume combustion of several fuel–air mixtures, in various conditions (initial pressures within 0.3–1.2 bar and ambient initial temperature) are reported. Experiments were performed in a spherical and three cylindrical vessels, with various diameters to height ratios. The measured explosion pressures will be examined versus the corresponding adiabatic values, calculated in the assumption that the chemical equilibrium is reached within the flame. One of the examined systems, propene–air, was already studied and results on its explosion in closed vessels were previously discussed in [5,7]. Some measurements were repeated in the present study, by means of a more performing acquisition system; only the new results are presented here.

2. Experimental

A vacuum and gas-feed line forms the experimental set-up, tight at pressures from 0.1 mbar to 1.5 bar. The line interconnects the vacuum pump, the gas cylinders with fuel and air, the metallic cylinder for mixture storage and the explosion vessels. The vacuum pump maintains a vacuum of 0.1 mbar in the explosion vessel, after each experiment.

The fuel–air mixtures were obtained by the partial pressure method and used 24 h after mixing the components, at a total pressure of 4 bar.

Experiments were performed in four explosion vessels, tight at vacuum and at pressures up to 20 bar: vessel S – a spherical vessel with the radius \( R = 5 \text{ cm} \); vessel C1 – a cylinder with \( h = 15 \text{ cm} \) and \( \Phi = 10 \text{ cm} \); vessel C2 – a cylinder with \( h = 9.8 \text{ cm} \) and \( \Phi = 4.9 \text{ cm} \); vessel C3 – a cylinder with \( h = \Phi = 6 \text{ cm} \). The initial pressures of fuel–air mixtures were measured by a strain gauge manometer (Edwards type EPS-10BM).

Ignition was made with inductive–capacitive sparks produced between stainless steel electrodes and the spark gap was located in the geometrical center of each vessel. Vessel C1 was fitted with a supplementary pair of electrodes, able to produce sparks 5 mm below the center of the upper lid. Each vessel was equipped with an ionisation probe used to monitor the arrival time of the flame front. Its tip was usually mounted 3 mm away from the sidewall.

The pressure variation during explosions was recorded with a piezoelectric pressure transducer (Kistler 501A), connected to a Charge Amplifier (Kistler 500/SN). The signals of the ionisation probe amplifier and of the Charge Amplifier were recorded with an acquisition data system TestLab™ Tektronix 2505, by means of an acquisition card type AA1, usually at 10⁴ signals/s. The Charge Amplifier was calibrated by means of a Kistler Calibrator type 5357.

Vessels C2 and C3 had a transparent window made from synthetic glass (3 cm diameter) mounted in the center of the upper lid, which enabled the visual observation of flame appearance and propagation.

Gaseous fuels: methane (99.5%), propene (99.5%), \( n \)-butene (99.0%), \( n \)-butane (99.0%), butadiene (99.5%) and pyrolysis gas (28.6% propane; 44.5% propene; 9.38% propyne; 8.22% allene and 8.33% butadiene) were purchased from Pitesti and Brazi Petrochemical Plants. Liquid fuels: \( n \)-pentane and \( i \)-pentane, \( n \)-hexane, cyclohexane and benzene (analytical grade) were used without a further purification.

Air was dried by means of a line containing \( \text{H}_2\text{SO}_4 \), KOH (s), \( \text{CaCl}_2 \) and silicagel with moisture indicator. The experimental procedure consists of evacuating the explosion vessel to 0.1 mbar; the fuel–air mixture is then introduced, at the desired pressure and it is allowed to become quiescent, then it is ignited and the signals of the acquisition system are captured, stored and evaluated. Minimum three experiments were performed for each initial condition of explosive mixture. For
a few systems (e.g. stoichiometric propene–air), several sets of 15 experiments were conducted in identical conditions, in the spherical vessel with central ignition. The standard error in measured explosion pressures was $\leq 2\%$.

Fuels which are liquid at ambient temperature and pressure were vaporised before mixing with air, by injecting the liquid into a spherical bulb evacuated down to 0.1 mbar, through a side tube having a vacuum tight injection septum at its end.

3. Computing programs

The calculations of adiabatic explosion pressures were made with the program ECHIMAD [24], based on a general algorithm meant to compute the equilibrium composition of products for any fuel-oxidizer gaseous mixture. The algorithm is based on the thermodynamic criterion of chemical equilibrium: the minimum of free Gibbs energy, at constant temperature and pressure or minimum of free Helmholtz energy, at constant temperature and volume. Fifteen compounds, among them one solid compound ($C_\text{graphite}$) were considered as products: the fuel ($C_n H_m$), $C_\text{graphite}$, $C O_2$, $CO$, $H_2O$, $O_2$, $N_2$, $C H_4$, $C_2H_2$, $C_2H_4$, $H_2$, $NO$, $H$, $OH$ and $O$). Their heat capacities (expressed as function of temperature with the form: $C_P = a + b \cdot T + c \cdot T^2 + d \cdot T^{-2}$), the standard enthalpies of formation at 298 K and the standard entropies at 298 K were taken from references [47,48]. For few substances not included in these data collections, the NASA polynomials [49] were used to calculate the heat capacities at various temperatures, data fitted then to the desired form given above.

Adiabatic flame temperatures were also calculated, for each system, both in isobaric and isochoric combustion. The constant pressure adiabatic flame temperatures of $CH_4$–air mixtures with various methane concentrations were used to compare the results obtained from the program ECHIMAD with those obtained with the Lite version of the program EQS4WIN, available from www.mathtrek.com [25]. The agreement was excellent, as shown in another recent publication [50].

4. Results and discussion

Methane–air is among the most studied systems, since methane is the main component of natural gas and coal mine gas. Because of this overflow of information, the present paper reports just a few data obtained in a series of experiments performed in cylindrical vessel C2, at initial pressures between 0.3 and 1.1 bar and methane concentrations within the lower flammability limit and the stoichiometric composition. A plot of explosion pressures attained in experiments at ambient initial pressure versus methane concentration is given in Fig. 1. In the same figure we also plotted results recently reported by Pekalski et al. [19] from measurements in a 20 L spherical vessel with central ignition [19], together with calculated explosion pressures by means of the program ECHIMAD and extracted from the already mentioned paper [19], using the CHEMKIN code and two reaction mechanisms [51,52]. The experimental results seem to fit the same curve, in spite of the great difference in the volume of the two explosion vessels. At the same time, quite small differences are observed between calculated explosion pressures, by using the two subroutines. As expected, the calculated adiabatic explosion pressures are systematically higher than measured values. Their values come closer at concentrations near the stoichiometric one, but show large deviations in the vicinity of the lower flammability limit.

The maximum explosion pressure from our measurements is 8.6 bar, obtained at [[$CH_4$]$=10.0\%$. Other values of measured maximum explosion pressures, collected from literature, are given in Table 1, together with the experimental conditions.

<table>
<thead>
<tr>
<th>No.</th>
<th>$P_{exp}$ (bar)</th>
<th>[%CH$_4$ (%vol)]</th>
<th>Explosion vessel</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>8.3</td>
<td>10.5</td>
<td>20L, according to EN 13673-1</td>
<td>[1,43]</td>
</tr>
<tr>
<td>2</td>
<td>8.1</td>
<td>9.5</td>
<td>20L, conditions as in EN 13673-1</td>
<td>[18]</td>
</tr>
<tr>
<td>3</td>
<td>8.5</td>
<td>10.9</td>
<td>20L, conditions as in EN 13673-1</td>
<td>[19]</td>
</tr>
<tr>
<td>4</td>
<td>8.3</td>
<td>10.0</td>
<td>20L, Sphere</td>
<td>[14]</td>
</tr>
<tr>
<td>5</td>
<td>8.2</td>
<td>10.2–10.5</td>
<td>20L, Sphere</td>
<td>[11,13]</td>
</tr>
<tr>
<td>6</td>
<td>8.1</td>
<td>9.5</td>
<td>5L, sphere</td>
<td>[9]</td>
</tr>
<tr>
<td>7</td>
<td>8.7</td>
<td>9.5</td>
<td>5L, sphere</td>
<td>[10]</td>
</tr>
<tr>
<td>8</td>
<td>7.7</td>
<td>9.5</td>
<td>204 m$^3$, Sphere</td>
<td>[30]</td>
</tr>
<tr>
<td>9</td>
<td>7.0</td>
<td>10.0</td>
<td>4.2L, sphere</td>
<td>[15]</td>
</tr>
<tr>
<td>10</td>
<td>8.0</td>
<td>9.5</td>
<td>20L, Sphere</td>
<td>[4]</td>
</tr>
</tbody>
</table>
Fig. 2. Measured and calculated explosion pressures of C₃H₆–air mixtures; $P_0 = 1$ bar; $T_0 = 298$ K. (■) Calculated, ECHIMAD; (□) spherical bomb; (●) cylindrical vessel C₁; (▲) cylindrical vessel C₂.

The maximum explosion pressures are $(8.3 ± 0.3)$ bar for measurements made in the 20L spherical vessel with central ignition; higher deviations are observed, however, when using different size vessels, especially larger ones, where radiative and convective heat losses to the walls are important. Our measurements, made in a much smaller vessel, are close to these data within an acceptable deviation.

Another fuel, extensively characterized by our computations and experiments in several closed vessels, is propene. Propene–oxygen and propene–air were less studied until now, when measurements performed by Pekalski et al. in a 20L spherical vessel and computations of explosion pressures [19] complete earlier published data [5,7].

The explosion pressures calculated by means of ECHIMAD code reveal the same good agreement with data calculated by CHEMKIN and reported by Pekalski et al. [19]. In the vicinity of stoichiometric concentration, the results from ECHIMAD are slightly higher as compared to those computed by CHEMKIN (relative error: 0.3–1.2%), but they are essentially the same in lean and rich mixtures. Our calculations were also extended up to 14% C₃H₆ and allowed the observation of soot appearance starting from 12.3%.

The measured explosion pressures for this system are greatly influenced by the vessel shape and volume, as seen from Fig. 2, where data measured in the spherical vessel S and in cylindrical vessels C₁ and C₂ were plotted together with the calculated explosion pressures.

For all investigated concentration range, the values measured in the 0.7L spherical vessel are close to the values measured in the 20L vessel, reported in [19]. The differences between the reported values can be explained by the differences in the dimension of the two vessels, as the vessel recommended in the standard procedure has a 25 times larger volume than spherical vessel S.

The maximum explosion pressures are 8.6 bar in vessel C₂, 9.0 bar in vessel C₁ and 9.6 bar in vessel S, observed at concentrations slightly greater than stoichiometric, between 4.8 and 5.5 vol.%. Other values of measured explosion pressures from literature are quite close: 9.2 bar [18] and 9.4 bar [19], both from measurements in 20L spherical vessels.

The calculated and measured explosion pressure curves versus propene concentration display a shape difference similar to the one observed by Pekalski et al. [19]: the calculated explosion pressures decrease very slowly when propene concentration increases from 4.5 vol.% (stoichiometric concentration) to the upper explosion limit (11.2 vol.% [46]), whereas the measured explosion pressures decrease almost symmetrically when [C₃H₆] approaches the lower and the upper explosion limit. As seen in Fig. 2, each set of measured explosion pressures versus propene concentration is well fitted by a second-order polynomial.

In some of our experiments, the visual observation of explosion (in vessels C₂ and C₃) allowed the determination of the soot threshold of propene in air at ambient initial pressure and temperature: 7.6%, a value confirmed by measurements performed on flames stabilized on a Bunsen burner by Takahashi and Glassman [53]. In opposition to this, the cited reference [19] indicated 12% as threshold of soot formation in propene–air mixtures, determined from the slope change observed in the plot of calculated explosion pressures versus [C₃H₆] [19], a value which is in fact higher than the upper flammability limit of this fuel, at ambient initial pressure and temperature.

For all propene–air mixtures, linear correlations were found between explosion pressures and the total initial pressure. Two sets of results obtained in spherical vessel S and in the cylindrical vessel C₁ are given in Figs. 3 and 4. In Fig. 3, only data referring to lean mixtures were plotted, but similar correlations were observed for rich mixtures as well. In Fig. 4, a comparison between calculated and measured explosion pressures is once more given. This time, the plots $P_{\text{exp}} = f(P_0)$ are drawn for explosions in the spherical vessel S and in the cylindrical vessel C₁, when ignition was made in the center of the vessel and near the top lid, respectively. The lowest explosion pressures were obtained after the asymmetric ignition, which determines maximum heat loss during propagation. The differences between
the explosion pressures measured at central and side ignition increase when the fuel content is closer to the most reactive concentration, where the combustion delivers the maximum amount of heat.

The explosion times \( \theta_{\text{exp}} \) in the spherical vessel \( S \) and cylindrical vessels \( C_1 \) and \( C_2 \) are greatly influenced by the vessel’s volume and by propene concentration (Fig. 5); within experimental errors, \( \theta_{\text{exp}} \) measured in cylindrical vessel \( C_2 \) were the same as \( \theta_{\text{exp}} \) measured in the spherical vessel. As expected, at constant \([C_3H_6]\), the highest explosion times are required by the largest volumes.

An earlier analysis of heat balance during the constant volume combustion of a gaseous fuel–air mixture [54] has shown that the explosion pressure \( P_{\text{exp}} \) can be correlated with the total initial pressure \( P_0 \) by the equation:

\[
P_{\text{exp}} = k_{\text{ad,V}} \cdot P_0 - q_{\text{tr}} \cdot \frac{\gamma_e - 1}{V_0}
\]

where \( k_{\text{ad,V}} \) is the adiabatic coefficient of the pressure increase during explosion, defined as \( \frac{P_{\text{ad}}}{P_0} \); \( q_{\text{tr}} \) the total amount of heat losses, transferred by the gas before the end of combustion; \( \gamma_e \) is the adiabatic coefficient of the burned gas, at the end of combustion; \( V_0 \) is the vessel volume.

Indeed, the data plotted in Figs. 3 and 4 are linear correlations with the general form:

\[
P_{\text{exp}} = a \cdot P_0 - b
\]

Such dependencies were found for all examined fuel–air mixtures, in all vessels. A set of data which represent \( b \), the intercept of such plots, determined in closed vessels \( S \) and \( C_1 \) for mixtures with variable propene concentration are given in Fig. 6(a and b).

As the adiabatic coefficient of burned gas \( \gamma_e \) has a moderate variation (from 1.250 to 1.230) when the propene concentration increases from 3 to 8%, the variation of \( b \) is entirely determined by the variation of the transferred heat \( q_{\text{tr}} \) towards the explosion vessel. The most reactive mixtures, having a fuel concentration close to the stoichiometric one, burn quickly and have less time left for heat losses as compared to lean and rich mixtures. Thus,
the increase of the explosion time allows the increase of heat transferred to the vessel, before the end of combustion. At constant propene concentration, the values of \( b \) determined in the spherical vessel are always lower as the corresponding \( b \) values determined in the cylindrical vessel C1, where the flame has contact with the side wall much earlier than with the top and bottom of the vessel. The values of \( b \) from cylindrical vessels C2 and C3 are simultaneously influenced by \( q_{tr} \) and \( V_0 \), so that a comparison with data plotted in Fig. 6(a and b) is not possible.

One has to observe that \( b \) values given in Fig. 6(a and b) are scattered; the same was found for experiments in cylindrical vessels C2 and C3. A possible explanation is the asymmetry of spark generation between the electrode tips: few sparks are really produced at mid-distance between the electrodes and thus each flame starts growing slightly deviated from the vessel’s center.

In a few experiments, explosions of pyrolysis gas–air mixtures were studied, in explosion vessel C1. The explosion pressures and explosion times measured at ambient initial conditions are plotted against fuel concentration in Fig. 7. The values of maximum explosion pressure and of explosion times are quite close to similar values determined for propene–air in vessel C1: the maximum explosion pressure was 8.5 bar and the minimum value of explosion time is 32 ms, recorded at 5.2–5.3% fuel. As the pyrolysis gas is a complex mixture of 3-carbon atoms hydrocarbons from several classes, it is not possible to ascertain this composition to some value of equivalence ratio.

The explosion pressures of several other hydrocarbon–air mixtures are given in Figs. 8–10. Few data from literature were available for comparison; only measurements for \( n \)-pentane–air in two spherical vessels with 0.5 and 4 L volumes were found and plotted together with our measurements, in Fig. 9.

Measured explosion pressures for \( i \)-pentane were, within experimental errors, identical to those for \( n \)-pentane and were not plotted any more. For C4- and C6-hydrocarbons both measured and calculated explosion pressures were plotted versus their equivalence ratio \( \phi \), for better comparison (Figs. 8 and 10). The best fits of explosion pressures against \( \phi \), calculated according to second-order polynomials, were also plotted for all data. The computed adiabatic explosion pressures are practically the same
for \( n-C_6H_{14} \)-air and \( i-C_6H_{14} \)-air and have close values to those characteristic to \( C_7H_{16} \)-air mixtures. At all equivalence ratios, the adiabatic explosion pressures were higher as compared to measured ones. Closer values of adiabatic and experimental explosion pressures were observed only for lean benzene-air mixtures. For other examined systems, the computed explosion pressures were higher than measured explosion pressures, especially for rich mixtures. At ambient initial conditions, the experimental maximum explosion pressures of these fuels range between 8.4 and 8.8 bar. They are obtained at equivalence ratios \( 1.0 < \phi < 1.2 \). A similar trend is noticed in the variation of explosion times with the equivalence ratio, for these mixtures. An illustration of their behavior is given in Fig. 11.

A comparison of the maximum explosion pressures reported in the present paper and recently reported values from literature, obtained only in spherical vessels of 20 L volume, are given in Table 2. For most fuels, the maximum explosion pressures measured in a 0.7 L sphere are indeed lower as compared to those obtained by using a 20 L sphere. However, differences are observed even between data measured by the same technique, as seen in Table 2 for methane-air and propene-air mixtures (data reported in references [18,19]). In such conditions, even the measurements made in smaller bombs (spherical or cylindrical) can be useful safety data for gaseous fuel-air mixtures.

<table>
<thead>
<tr>
<th>Fuel</th>
<th>Present paper</th>
<th>Reference [18]</th>
<th>Reference [19]</th>
</tr>
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<tr>
<td>Methane</td>
<td>8.6</td>
<td>8.1</td>
<td>8.5</td>
</tr>
<tr>
<td>( n )-Pentane</td>
<td>8.4</td>
<td>9.5</td>
<td></td>
</tr>
<tr>
<td>( n )-Hexane</td>
<td>8.7</td>
<td>9.5</td>
<td></td>
</tr>
<tr>
<td>( c )-Hexane</td>
<td>8.7</td>
<td>9.4</td>
<td></td>
</tr>
<tr>
<td>Propane</td>
<td>9.4</td>
<td>9.4</td>
<td>9.2</td>
</tr>
<tr>
<td>Propene</td>
<td>8.5</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>( n )-Butane</td>
<td>8.5</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>( i )-Butene</td>
<td>8.6</td>
<td>7.9</td>
<td></td>
</tr>
<tr>
<td>Benzene</td>
<td>8.8</td>
<td>9.8</td>
<td>–</td>
</tr>
</tbody>
</table>

5. Conclusions

Measurement of explosion pressures of several fuel-air mixtures, at various initial pressures, in closed vessels of different shapes and volumes has shown a similar behavior of investigated fuels: methane, \( n \)-pentane, \( n \)-hexane, propene, \( i \)-butene and \( i \)-butene; cyclohexane, butadiene and benzene. Linear correlations were found between explosion pressures and initial pressures of fuel-air mixtures, for sets of experiments made in a closed vessel, at constant initial temperature. The slope of these correlations are close to the adiabatic explosion pressure; their intercept depends both on the heat lost by the burning mixture to the vessel and on vessel’s volume.

The measured explosion pressures were compared with the adiabatic explosion pressures, computed assuming that chemical equilibrium is reached in the flame front. Due to inherent heat losses during flame propagation, experimental explosion pressures are lower than adiabatic values. The heat losses are higher in cylindrical vessels as compared to a spherical vessel, even when ignition is performed in the geometrical center of these vessels.

The explosion pressures depend on fuel concentration and reach their maximum value in the field of rich mixtures, with an equivalence ratio \( \phi = 1.1–1.3 \). Experimental data were fitted against fuel concentration or equivalence ratio by second-order polynomials. The fitted curves can be useful for finding explosion pressures by interpolating the actual measured values.

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