

Pressure evolution of ethylene-air explosions in enclosures

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Abstract. The peak explosion pressure and the maximum rate of pressure rise are important safety parameters for assessing the hazard of a process and for design of vessels able to withstand an explosion or of their vents used as relief devices. Using ethylene-air with various fuel concentrations (4-10 vol% C₂H₄) as test mixture, the propagation of explosion in four closed vessels (a spherical vessel with central ignition and three cylindrical vessels with various L/D ratios, centrally or side ignited) has been studied at various initial pressures between 0.3-2.0 bar. In all cases, the peak pressures and the maximum rates of pressure rise were found to be linear functions on the total initial pressure, at constant fuel concentration. Examining several enclosures, the maximum values of explosion pressures and rates of pressure rise have been found for the spherical vessel. For the same initial conditions, the peak explosion pressure and maximum rates of pressure rise determined in cylindrical vessels decrease with the increase of L/D ratio. Asymmetric ignition, at vessel's bottom, induces important heat losses during flame propagation. This process is characterized by the lowest rates of pressure rise, as compared to propagation of flame ignited in the centre of the same vessel.

1. Introduction

Explosions of fuel-air gaseous mixtures in enclosures are characterized by specific characteristic parameters: the peak explosion pressure, the time to peak explosion pressure, the maximum rate of pressure rise, the propagation speed and the normal burning velocity [1]. The explosions may take place as deflagrations (subsonic propagation speeds) or detonations (larger than sonic propagation speeds). Deflagrations develop usually high explosion pressures (6-9 times the initial pressure), reached in a short time ranging from a 20-50 ms (in small vessels, with volumes below 1 L) to a few seconds (in vessels with volume between 1 L and a few m³). During such processes the rate of pressure rise varies between 100 and 2000 bar/s, depending on initial composition, pressure and temperature of the flammable mixture, on vessel's form and volume, on ignition source position and energy. Knowledge of characteristic parameters of deflagrations and of factors that influence them allows the assessment of explosion risks for flammable mixtures in various conditions and the formulation of safety recommendations against the damaging effect of such explosions. At the same time, the characteristic parameters of deflagrations are necessary input data for the design of venting systems and for modelling the flame propagation in various conditions. Explosion pressures and maximum rates of pressure rise in closed vessel explosions are influenced by the initial composition, pressure and temperature of the fuel-air mixture (factors which determine the rate of heat release) and



by the volume and the shape of the enclosure, the ignition source size, energy and position, the pre-existing or combustion-created turbulence (factors which determine the amount of generated heat as well as the amount of heat losses during flame propagation)[1-3]. Specific propagation features appear when the process takes place in elongated vessels, such as the preheating enclosures for chemical reactors or the pipes connecting fuel tanks. Three stages during flame propagation in long closed vessels have been observed [2]; the flame configuration, propagation speed and aspect of $p(t)$ evolution have specific features in each period. The first stage is characterized by a quasi-spherical flame propagation, accompanied by maximal propagation speeds. Here, the flame is accelerated due to the expansion of burned gas and the flame structure is still uniform and homogeneous. At the end of the first stage, the flame front is stretched in axial direction and it is slowed down, as a consequence of decrease in its surface at the contact between flame and vessel's walls. Studies on flame propagation of propane-air mixtures in elongated vessels with L/D ranging between 1 and 19 [3] showed that the pressure increase is almost linear and the propagation speed is quasi-constant in the 2nd stage, depending on the equivalence ratio of the flammable mixture and on vessel's geometry. In the last stage, the propagation speed was again decreasing and the flame instability increased so that the amplitude of peak-to-peak pressure oscillations could reach 5 bar. Other specific features of fuel-air deflagrations in tubes with asymmetrical ignition have been outlined by works of Phylaktou et al. [4,5] Fairweather et al. [6], Kindracki et al. [7], Cammarota et al. [8], Xiao et al. [9,10]. Bielert et al. [11] developed a one-dimensional model to simulate the combustion of methane-air flames in long closed pipes ($L/D = 12.3$ or 18.4), including the heat transfer to walls into simulation. Numerical simulations of fuel-air flames propagating in pipes have been performed also by Bychkov et al. [12] for propane-air flames (using data from [13]) and Bi et al. [14] for methane-air flames (using data from [7]). Other recent publications [10,15,16] examine characteristic aspects of flame propagation in tubes, relevant for the appearance of characteristic tulip flame after the rapid decrease of flame area due to the flame quenching at walls. Flame propagation in long vessels has been investigated also in connection with the process of DDT (Deflagration-to-Detonation Transition) frequently met in such conditions [17].

Ethylene, widely used in the chemical industry for production of valuable products (ethylene oxide, ethylene dichloride or polyethylene), was chosen as test fuel due to fire and explosion hazards associated to its mixtures with air, oxygen or other oxidizers. Explosions of gaseous ethylene-air mixtures in enclosures have been studied in various conditions: in spherical and cylindrical vessels, at initial pressures within 1 - 20 bar and initial temperatures within 20 - 200° C, using mixtures with variable ethylene concentration [4,18-21]. The propagation of C_2H_4 -air flames in a cylindrical vessel with $L/D = 21.6$ and side ignition was studied by Phylaktou [4] who reported the rates of pressure rise and the flame speeds in various stages of flame propagation. Other results on ethylene-air explosions in closed vessels (explosion pressures and severity factors at variable initial pressures (1 - 10 bar), initial temperatures (20 - 200 °C) and C_2H_4 concentration (3 - 30 vol.%) have been recently obtained in the frame of European Safekinex project [20]. Ethylene-air explosions in cylindrical vessels with L/D between 1.0 and 2.4 and central ignition have been experimentally investigated at ambient temperature and various initial pressures within 0.2 and 1.1 bar [21].

The present paper reports maximum (peak) explosion pressures, explosion times, maximum rates of pressure rise and severity factors reached during asymmetrically ignited explosions of ethylene-air in closed cylindrical vessels with L/D between 2.4 and 20.7 in comparison with centrally ignited explosions in elongated vessels and a spherical vessel. All data are discussed in correlation to the initial pressure and composition of ethylene-air mixtures and with the characteristic features of closed vessels: volume V and aspect ratio L/D .

2. Experimental

The main components of the experimental set-up are the explosion vessels, the cylinder for gaseous mixture storage, the ignition controller and the acquisition data system. The set-up has been described in previous papers [21,22]. Several explosion vessels, with different L/D ratios, have been used in the present measurements. The vessels were made from stainless steel and were equipped with several

ports for the gas feed/evacuation valve and for mounting the ionization probes, thermocouples, pressure transducers and ignition electrodes. Each vessel was rated to 40 bar. The mixtures were ignited in vessel's centre or near the bottom (in the case of cylindrical enclosures), at points axially situated along each vessel, using high voltage inductive-capacitive sparks. Their list and characteristic dimensions are given in Table 1.

The transient pressure variation was recorded by means of two piezoelectric pressure transducers (Kistler 601A), in line with Charge Amplifiers Kistler 5001 SN. One pressure transducer was mounted in the centre of the top cover; the other was mounted in the bottom flange, flush with the cylindrical wall, 15 mm far from bottom. The time of flame arrival to the top was monitored by means of 2 ionization probes: one of them was mounted flush with the cylindrical wall, 15 mm far from bottom, the other in a symmetrical position relative to the top cover. The signals from the 2 ionization probes and from the 2 Charge Amplifiers were acquired, at 2500 signals/channel and a maximum of 1 GS/s sampling rate, with a 4-channels Tektronix digital oscilloscope type TDS 2014B connected to a PC.

Table 1. Geometric characteristics of explosion vessels.

Vessels		L / cm	D / cm	V / cm ³	L/D
Spherical	S	-	10.0	524	-
	V1	15.0	10.0	1178	1.5
Cylindrical	V2	20.0	8.1	1030	2.4
	V3	54.7	6.8	1986	8.1
	V4	100.0	4.8	1809	20.7

The following operating procedure has been used: 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 initiating spark was used as external trigger of the time base of the oscilloscope.

Ethylene, purity 99.99% (SIAD - Italy) and compressed air, has been used without further purification. Experiments have been made on ethylene-air mixtures with various concentrations within 3.0 - 10.0 vol% at initial pressures within 0.30 - 1.8 bar and ambient initial temperature. Minimum 3 experiments have been performed for each initial condition of explosive mixture.

3. Data evaluation

The computation of maximum explosion pressures from $p(t)$ diagrams was made after smoothing the data either by FFT method or/and by using a cubic spline function. The rates of pressure rise were computed by numerical derivation of smoothed $p(t)$ data using the 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. In all cases, we used a 10% smoothing level, since higher values of this level (e.g. 20%) leads to a reduction of the noise accompanied by the signal distortion.

The calculation of adiabatic explosion pressures and flame temperatures of ethylene-air mixtures at various initial pressures has been made with the program COSILAB [23] based on a general algorithm meant to compute the equilibrium composition of products for any fuel–oxidizer–diluent gaseous mixture. The algorithm is based on the thermodynamic criterion of chemical equilibrium: the minimum Gibbs energy, at constant temperature and pressure or the minimum Helmholtz energy, at constant temperature and volume. Fifty-three compounds have been considered as products.

4. Results and Discussion

Typical plots of pressure–time history for explosions propagating in several cylindrical vessels are shown in Figures 1 (central ignition) and 2 (bottom ignition), for the stoichiometric ethylene–air mixture ($[C_2H_4] = 6.54$ vol %) at ambient initial pressure. In Figure 1, the $p(t)$ data measured in the spherical vessel with central ignition, for the same ethylene–air mixture, are overlaid. In centrally

ignited explosions (Figure 1), the shortest explosion time (time from ignition to the peak explosion) is reached in the spherical vessel. In this vessel, both the peak explosion pressure and the maximum rate of pressure rise exceed the corresponding indices measured in the cylindrical vessels. A common feature of $p(t)$ records from cylindrical vessels is the appearance of a short period of rapid pressure increase, at the beginning of flame propagation. In this stage, the pressure rise of the examined explosions increases according to the cubic law ($\Delta p = k \cdot t^3$) [24] so one can assume that the flame propagates undisturbed according to a spherical symmetry. As the L/D ratio of vessels increases, the duration and height of the rapid pressure increase are lower. In the second stage of both central and the bottom ignitions, the explosions are characterized by lower pressure rise rates and longer explosion times. A set of relevant sets of explosion indices is given in Table 2, where θ_{max} is the time to maximum explosion pressure. In accord to Phylaktou et al. [4] we use the terms “the first rate of pressure rise” and “the second rate of pressure rise” for the pressure rise rate of the first and the second stage of propagation, respectively.

Smooth $p(t)$ records have been obtained in centrally ignited explosions (Figure 1). Bottom-ignited explosions in vessels with $L/D > 2.4$ (Figure 2) are accompanied by pressure oscillations that appear in the last stage of propagation, when flame instability develops. The oscillations are observed both in lean and rich mixtures; their amplitude and frequency depend on the equivalence ratio and initial pressure of flammable mixtures and L/D values of explosion vessels. The peak-to-peak amplitude of oscillations recorded in vessel V4 reached 2.2 bar during the explosion of a 5% C_2H_4 -air mixture ($\varphi=0.756$) at $p_0 = 1$ bar. In a shorter vessel, the amplitude of oscillations reached 2.3 bar during the explosion of the stoichiometric C_2H_4 -air mixture. In all cases, the pressure oscillations create a more dangerous situation of experiments in comparison with oscillation-free runs.

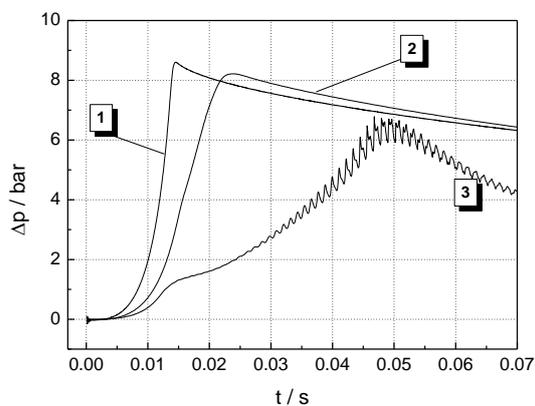


Figure 1. Pressure–time history for explosions of the stoichiometric C_2H_4 -air mixture at $p_0 = 1$ bar in vessels with central ignition: (1) vessel S; (2) vessel V1 ($L/D = 1.5$); (3) vessel V3 ($L/D = 8.1$).

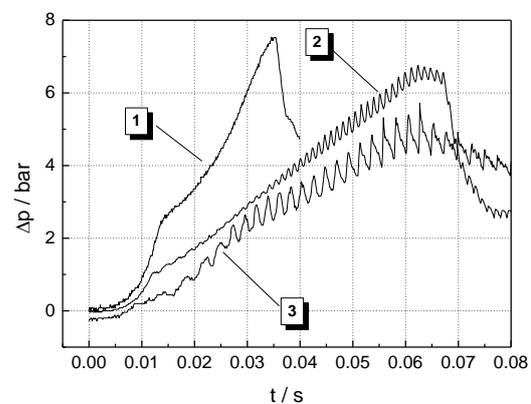


Figure 2. $p(t)$ for explosions of the C_2H_4 -air stoichiometric mixture at $p_0 = 1$ bar; vessels with bottom ignition: (1) V2 ($L/D = 2.4$); (2) V3 ($L/D = 8.1$); (3) V4 ($L/D = 20.7$)(offset -0.2 bar).

4.1. The maximum explosion pressures

As in the case of centrally ignited explosions studied in spherical vessel S and in cylindrical vessel V1 using other gaseous fuel-air mixtures (CH_4 , C_3H_8 , C_5H_{12} , C_3H_6 , C_4H_6 or C_6H_6 [21,22,25-27]) a linear correlation was found between the peak (maximum) explosion pressure p_{max} and the total initial pressure p_0 :

$$p_{max} = m \cdot p_0 - n \quad (1)$$

The correlation holds for all vessels and all fuel concentrations where oscillation-free explosions occur, in deflagration regime. Data relevant for explosions of three ethylene-air mixtures in vessel V3 are given in Figures 3 and 4. The peak pressures reached in bottom-ignited explosions are lower as

compared to the peak pressures reached in centrally ignited explosions, over the whole range of initial pressures.

Table 2. Deflagration indices measured during explosions at $p_0 = 1$ bar of the stoichiometric (6.54 vol.%) C_2H_4 -air mixture in several vessels with central ignition.

Vessel's name	L/D	p_{max} / bar	1 st rate of pressure rise $(dp/dt)_{max1}$ / (bar s ⁻¹)	2 nd rate of pressure rise $(dp/dt)_{max2}$ / (bar s ⁻¹)	θ_{max} / ms
S	-	9.63	2960	-	14
V1	1.5	9.18	820	-	16
V2	2.4	6.16	734	814	20.5
V3	8.1	7.28	268	300	46.8
V4	20.7	6.33	79	105	58.0

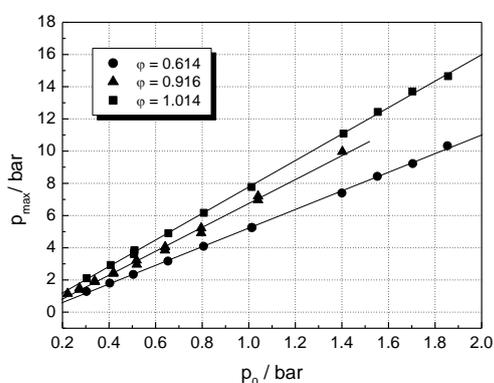


Figure 3. Maximum explosion pressures reached in vessel V3 (L/D = 8.1) with central ignition; C_2H_4 -air mixtures with various initial concentrations.

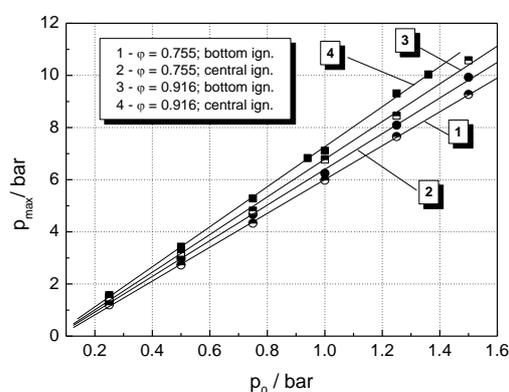


Figure 4. Maximum explosion pressures reached in vessel V3 (L/D = 8.1) with central and with bottom ignition; lean C_2H_4 -air mixtures.

The influence of the aspect ratio L/D on peak explosion pressures is shown in Figures 5 and 6, for bottom-ignited explosions; Similar diagrams have been drawn for centrally ignited explosions.

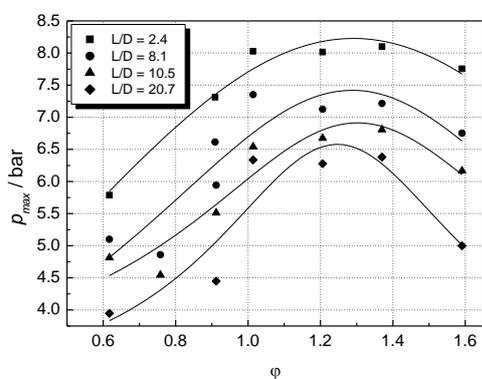


Figure 5. Influence of initial concentration on average peak explosion pressures at $p_0 = 1$ bar, in vessels with various L/D and bottom ignition.

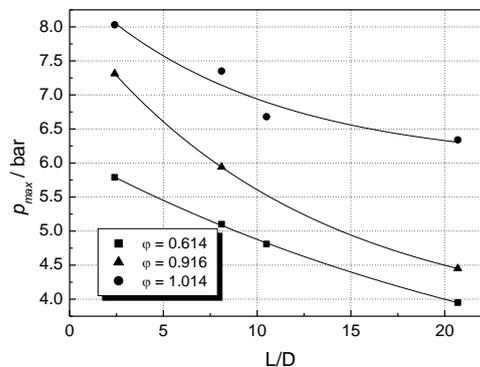


Figure 6. Influence of L/D on average peak explosion pressures of several ethylene-air mixtures at $p_0 = 1$ bar; bottom ignition.

For the stoichiometric and rich mixtures, characterized by appearance of important pressure oscillations in the 2nd and 3rd stages of propagation, the data scattering in Figures 5 and 6 is larger as compared to lean ethylene-air mixtures, since the peak explosion pressures have been evaluated as average values between the pressure peaks corresponding to pressure oscillations. For all vessels, the maximum explosion pressure is observed in the range of rich fuel-air mixtures ($[C_2H_4] = 7-8 \text{ vol}\%$). At constant composition, the increase of L/D results in the decrease of peak explosion pressures, due to a longer time of the heat exchange between the hot combustion products and the cold vessel walls.

4.2. The first and second rates of pressure rise

In all experiments, the first stage of flame propagation was oscillation-free and the $(dp/dt) = f(t)$ diagrams were easily evaluated. In the 2nd stage of experiments in vessels with $L/D > 2.5$, where pressure oscillations have been observed for all ethylene-air compositions, one could determine instantaneous second rate of pressure rise. Relevant information on the possible damage of explosions in elongated vessels is based, however, on the averaged second rate of pressure rise, computed from smoothed $p(t)$ data. Both the first and the second rates of pressure rise seem to follow linear correlations on initial pressure, p_0 . Typical results are plotted in Figures 7 and 8.

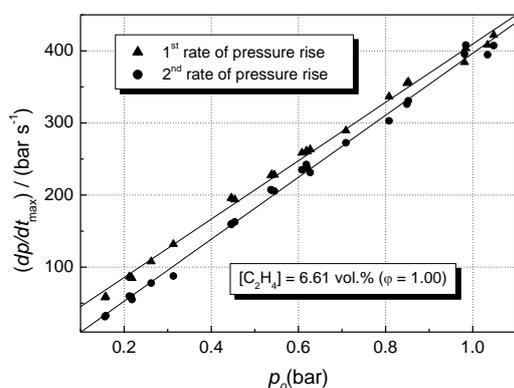


Figure 7. Initial pressure influence on maximum rates of pressure rise measured in vessel V2 ($L/D = 2.4$); bottom ignition.

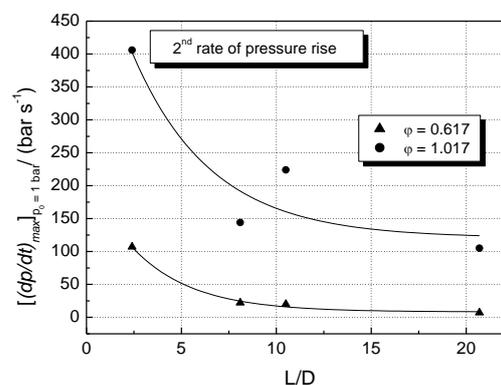


Figure 8. Influence of L/D on maximum rate of pressure rise measured for two ethylene-air mixtures at $p_0 = 1 \text{ bar}$; bottom ignition.

The influence of ethylene concentration on first and second rates of pressure rise is seen in Figure 9, where typical results from vessel V4 with bottom ignition have been plotted together with data from literature [4], measured in a horizontal tube with $L/D = 21.7$ and side ignition. Larger differences between the 1st and the 2nd rate of pressure rise were observed in this vessel, in comparison to other vessels.

The first rates of pressure rise of bottom ignited explosions have been used to calculate the severity factors characteristic for ethylene explosion in elongated vessels, as: $K_{G,L} = \left(\frac{dp}{dt}\right)_{\max} \cdot L$, according to a suggestion from Phylaktou [5]. Here, L (vessel's length), replaced the characteristic dimension $\sqrt[3]{V}$ used in the usual definition of a severity factor $K_G = \left(\frac{dp}{dt}\right)_{\max} \cdot \sqrt[3]{V}$. Representative

results are given in Fig. 10. For comparison, $K_{G,L} = 184 \text{ bar m s}^{-1}$ has been computed from literature data [4] (stoichiometric ethylene-air mixture, vessel with $L = 1.64 \text{ m}$, bottom-ignited explosion), a lower value in comparison to our data obtained in vessel V4 with a close aspect ratio, but lower length. Making use of the usual definition of the severity factor, we would obtain $K_G = 14.2 \text{ bar m s}^{-1}$ for the bottom-ignited explosion of the stoichiometric mixture in vessel V4.

4.3. Examination of heat losses in explosions propagating in elongated vessels with bottom ignition

The energy lost to the walls during explosion propagation in a closed cylindrical vessel has been estimated from the difference between the internal energy of burned gas in the ideal end condition of adiabatic combustion (characterized by $p_{max, ad}$ and $T_{max, ad}$) and in the real end condition (characterized by $p_{max, real}$ and $T_{max, real}$)[3]. According to this, the heat lost to the surface unit of wall,

q_{tr} , is: $q_{tr} = \frac{V}{A} \frac{1}{\gamma_{end} - 1} (P_{max, ad} - P_{max, real})$, where V and A are the volume and the inner surface area of the vessel, respectively, and is γ_{end} the adiabatic coefficient of burned gas, at flame temperature.

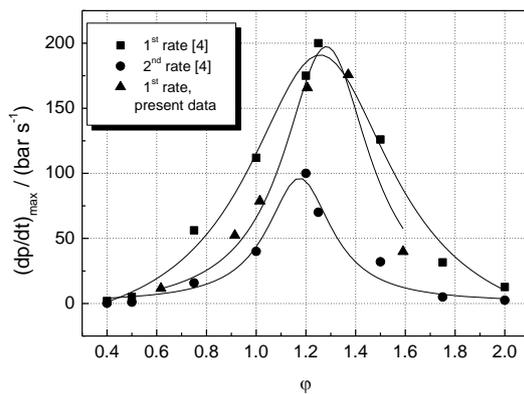


Figure 9. Maximum rates of pressure rise in vessels with bottom ignition, for mixtures with various concentrations at $p_0 = 1$ bar: (\blacktriangle) “First” rates, present data, vessel V4 ($L/D = 20.7$); (\bullet) “First” rates, vessel with $L/D = 21.6$ [4]; (\blacksquare) “Second” rates, as above [4].

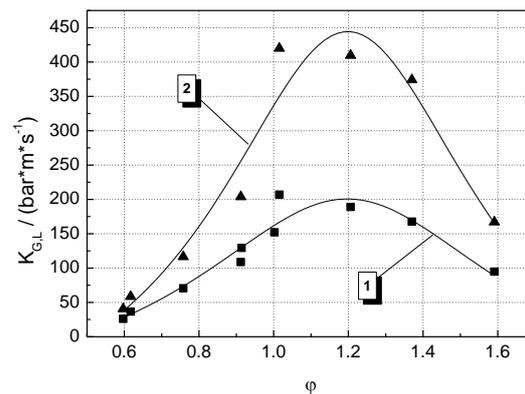


Figure 10. Severity factors of explosions at $p_0 = 1$ bar in elongated vessels with bottom ignition, for ethylene–air mixtures with various concentrations: (1) vessel V3 ($L/D=8.1$); (2) vessel V4 ($L/D=20.7$).

A set of data referring to explosions of ethylene-air mixtures at $p_0 = 1$ bar, computed for cylindrical vessels V2 - V4, is given in Table 3. The adiabatic coefficient of burned gas at flame temperature, γ_{end} , has been computed by means of burned gas composition obtained from equilibrium computations. The highest amount of transferred heat to the unit surface of walls was observed in vessel V4, characterized by the highest aspect ratio ($L/D = 20.7$) and the lowest - in vessel V2 ($L/D = 2.4$). For each case, the heat lost to wall's surface unit was found to depend linearly on the initial pressure.

Table 3. Heat lost to the surface unit of wall, $q_{tr} / (J m^{-2})$, during ethylene-air explosions at $p_0 = 1$ bar.

	[C ₂ H ₄] / vol%	6.62	7.77	8.73
L/D				
2.4		10.92	11.02	11.17
8.1		16.48	16.79	17.76
20.7		18.19	18.95	19.61

5. Conclusions

In elongated vessels, the deflagration indices are strongly influenced by the aspect ratio L/D of the vessel and reach much lower values as compared to short cylindrical vessels or with a sphere.

The important heat losses occurring in such situations explain the found dependencies of peak explosion pressure and of first and second rates of pressure rise against L/D . For all studied systems, linear correlations $p_{max} = f(p_0)$ and $(dp/dt)_{max} = f(p_0)$ have been found. The amount of transferred heat

to the unit surface of walls, computed from the adiabatic and experimental explosion pressures, had highest values for the vessel characterized by the highest aspect ratio ($L/D = 20.7$). The reported measurements provide useful results concerning explosion evolution in ethylene–air mixtures, in closed vessels with increasing asymmetry and might be useful for scaling explosions in chemical reactors, which are in most cases elongated cylindrical vessels.

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Acknowledgment: This work was supported by a grant of the Romanian National Authority for Scientific Research, CNCS – UEFISCDI, project PN-II-RU-PD-2012-3-0035.