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Additive effects on the rate of pressure rise for ethylene–air deflagrations in closed vessels

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HIGHLIGHTS

- Ethylene–air diluted by Ar, N_2 or CO_2 deflagrations were studied in closed vessels.
- The maximum rate of pressure rise $(dp/dt)_{max}$ and deflagration index K_G were reported.
- The influence of p_0, additive and geometry of explosion vessel were discussed.
- Linear correlations between $(dp/dt)_{max}$ and K_G versus p_0 were found.
- Adiabatic (maximum) deflagration indices were reported and discussed versus $K_{G,exp}$.

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ABSTRACT

The deflagration of stoichiometric ethylene–air mixtures without or with additives (Ar, N_2 or CO_2) in spherical and cylindrical closed vessels with central ignition was investigated in order to determine the maximum rates of pressure rise $(dp/dt)_{max}$ and the deflagration indices (severity factors) K_G. The influence of initial pressure, additive concentration, shape and size of explosion vessel on $(dp/dt)_{max}$ and K_G was studied. A linear correlation between the maximum rate of pressure rise and the total initial pressure was found for all examined systems, at constant initial temperature and fuel/oxygen ratio. The deflagration index K_G of centrally ignited explosions, calculated from experimental values of pressure rise rates, was compared with the adiabatic (maximum) deflagration index $K_{G,\max}$ calculated from the normal burning velocities and maximum explosion pressures of ethylene–air and ethylene–air-additive mixtures. The differences between experimental and adiabatic deflagration indices highlight the heat losses appearing in the last stage of explosions occurring in cylindrical vessels, strongly influenced by the length to diameter ratio, initial pressure and composition of flammable mixtures.

1. Introduction

The explosions occurring in domestic environments or chemical plants due to accumulation of combustible gases are frequent accidental events taking place during storage, transportation and manufacturing of gaseous fuels, failure of chemical and/or petrochemical plants, failure of high pressure vessels, fuel leaks in buildings, cleaning the fuel tanks of planes and ships. In process plants, explosive atmospheres may be formed inside the equipment (indoor) as a result of normal process conditions and outside due to abnormal processes and work practices as a result of failure of equipment protection and releases of flammable, explosive or oxidizing substances.

The evolution of a gaseous explosion in a closed vessel is characterized by the peak (maximum) explosion pressure, the time to peak pressure and the peak rate of pressure rise – parameters depending on the initial composition, pressure and temperature of flammable mixture, on volume and shape of the explosion vessel and on energy and position of the ignition source [1–3]. The peak (maximum) rate of pressure rise is further used for calculating the deflagration index (or "severity factor") K_G of gaseous explosions in enclosures according to the cubic-root law:

$$K_G = \left(\frac{dp}{dt}\right)_{max} \sqrt[3]{V} \tag{1}$$

K_G was defined by analogy to the deflagration index of dust-air explosions, K_{st}, introduced by Bartknecht and Zwahlen [4] and it was meant to normalize the maximum rate of pressure rise, largely dependent on vessel's size, with respect to the volume of the vessel. Knowledge of these flammability parameters is necessary for all

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Nomenclature

h	height (m)
K	deflagration index (bar m s^{-1})
m	pressure exponent of maximum rates of pressure rise (−)
p	pressure (bar)
r	vessel's radius (m)
s	speed, velocity (m s^{-1})
t	time (s)
t	temperature (K)
v	volume (m^3)

Greek

α, β	slope and intercept of $(dp/dt)_{max}$ vs. p_0
γ	adiabatic compression coefficient
μ	thermal coefficient of the normal burning velocity
ν	baric coefficient of the normal burning velocity
ρ	density (kg m^{-3})
Φ	diameter (m)

Subscripts

e	end (final) state of the burned gas
f	referring to a flame
G	referring to gas
max	maximum value
rel	relative value
St	referring to "Staub" (dust)
u	unburned gas
0	initial condition

fields of human activity where gaseous flammable mixtures are processed or transported, with the scope of assessing the potential damages produced by explosion in an enclosure and mitigating such damages. At the same time, the peak explosion pressure, the time to peak pressure and the peak rate of pressure rise are important input properties necessary for design of safe enclosures and relief vents.

Addition of diluents is one of the most effective ways to prevent damage from a deflagration in a closed vessel, by decreasing both the pressure rise and the maximum rate of pressure rise determined by the decrease of both evolved and sensible heat and of normal burning velocity [4–8]. Many experimental and numerical studies were focused on additive influence on flammability parameters of hydrogen–, methane–, propane–, LPG (Liquefied Petroleum Gas)–, ethylene–, propene–air mixtures [5–21]. Various inert diluents such as Ar and N_2, reactive diluents (CO_2, H_2O(vap) and/or exhaust gas, formed by the mixture of N_2, CO_2 and H_2O(vap)) and inhibitors (halogenated hydrocarbons) are among studied additives. Most data refer to additive influence on laminar burning velocity [6,12–14,17–21], as the burning velocity has a strong influence on the rate of heat release within a flame and on the rate of pressure rise during explosions in enclosures. Numerous studies report explosion pressures of fuel–air mixtures in the presence of various additives [5,7,8,10,11,14,15,17–19] and experimental measurements could be examined against adiabatic explosion pressures, computed after assuming that chemical equilibrium is established among combustion products in flames. The effort to predict the rates of pressure rise for closed-vessel explosions was less fruitful. Even if some authors could compute explosion pressures at various stages of flame propagation in a closed vessel [22–25], the time to reach the peak explosion pressure and/or the maximum rate of pressure rise are not yet available from propagation modeling by taking into account the heat losses at various stages of the process.

In many experimental and numerical studies on flammability parameters, ethylene has been used as a test fuel [6,8,13,21,26–37]. This interest can be explained by the wide use of ethylene as a raw material for ethylene oxide, ethylene glycol and polyethylene manufacture. Ethylene is among the key intermediates in the oxidation of higher hydrocarbons; this fact promoted the interest for numerical methods of kinetic modeling meant to find an adequate mechanism for ethylene oxidation, covering a wide range of temperatures, pressures, and equivalence ratios [16,33–37] by comparing the numerical predictions with experimental data obtained from high-pressure flow reactors, shock-tube studies of ethylene oxidation and pyrolysis and laminar premixed flames.

In the present work, previous studies on additive influence on flammability characteristics of ethylene–air explosions in enclosures [8,32] are continued. Data on maximum rates of pressure rise and deflagration indices are presented for stoichiometric ethylene–air-additive mixtures in three closed vessels: a spherical and two cylindrical vessels with different height to diameter ratios. The results are compared to those referring to the stoichiometric ethylene–air mixture. The study is focused on the influence of initial pressure and additive concentration on maximum rates of pressure rise and deflagration indices. The influence of the shape and the volume of the explosion vessel on these parameters are also examined.

2. Experimental

Experiments at ambient initial temperature and various initial pressures between 0.3 and 1.2 bar were performed in three closed vessels with central ignition: spherical vessel S (radius R = 5 cm), cylindrical vessel C1 (height h = 15 cm, diameter Φ = 10 cm) and cylindrical vessel C2 (h = Φ = 6 cm). Ignition was made with inductive capacitive sparks produced between two stainless steel electrodes. In each vessel, the pressure variation during explosion was recorded with a piezoelectric pressure transducer (Kistler 601A), connected to a Charge Amplifier (Kistler 5001 N); for detection of flame front position, an ionization probe was mounted in equatorial position.

Other characteristic features of experimental set-up have been presented in previous publications [7,8,20,21,32].

Ethylene–air and ethylene–air-additive mixtures were obtained by the partial pressure method, at a total pressure of 4 bar in a 10 L gas cylinder and were used 24 h after mixing the components. Ar and N_2 were used in a concentration range of 5–50 vol.%; CO_2 was used in a concentration range of 5–32 vol.%.

Ethylene (99.9%), Ar (99.99%), N_2 (99.99%) and CO_2 (99.5%) were purchased from SIAD, Italy and were used without further purification. Systems measured in the spherical vessel S and cylindrical vessel C1 were stoichiometric C_2H_4–air mixtures diluted with inert additives (Ar and CO_2). Systems measured in the cylindrical vessel C2 were stoichiometric C_2H_4–air mixtures diluted with Ar, N_2 and CO_2.

The experimental procedure consists of evacuating the combustion vessel down to 0.1 mbar; the fuel–air mixture was then introduced, allowed to become quiescent and ignited.

For a few systems, several sets of seven experiments were conducted in identical conditions in the cylindrical vessel C2 and the average error in measured explosion pressure was 2%.