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Additive influence on ignition of stoichiometric ethylene-air mixture by break sparks

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ABSTRACT

The minimum ignition currents for the stoichiometric ethylene-air mixture diluted with various amounts of argon, nitrogen or carbon dioxide were measured at various initial pressures within 20 and 120 kPa by using break sparks produced between Cd and W electrodes. The baric coefficients of ignition currents were determined from the correlation of minimum ignition currents with the total initial pressure. Compared to the previously reported ignition energies of initiation by high voltage inductive-capacitive sparks, the ignition energies of initiation by break sparks are systematically higher, due to important energy losses appearing at the contact between electrodes. From the minimum ignition currents of ethylene-air-additive mixtures, the maximum experimental safe gaps (MESG) were calculated using an empirical phenomenological correlation reported in literature. The measured minimum ignition currents, as well as the derived ignition energies and maximum experimental safe gaps are discussed in connection with the amount and nature of inert additive of ethylene-air mixtures.

1. Introduction

Explosions of flammable fuel-air gaseous mixtures represent a potential hazard in various industrial activities and daily life. In evaluating the explosion risk for gaseous flammable mixtures, consideration must be given to both components of the risk, i.e. the probability of explosion (bound to explosive atmosphere occurrence and to the probability of the presence of effective ignition sources) and the possible severity of consequences [1,2]. An explosive atmosphere is formed within the flammability range of any fuel-oxidizer mixture, characterized by the lower and the upper flammability limits (LEL and UEL), the limiting oxygen concentration (LOC) and the critical (minimum) inert concentration (CIC). Inside the flammability range, the fuel-air mixture may explode if it is brought into contact with a local source of energy (a hot body, an electric spark, a high energy electromagnetic radiation, a hot jet of burnt gases). In the absence of initiating source, the flammable mixture can be maintained indefinitely without runaway. The flammable mixtures can be brought outside the flammability range by variation of fuel/oxidant ratio, by decreasing the pressure or by adding inert components [3,4]. Outside the flammability range, the fuel-oxidizer mixture may burn, but the evolved heat is not sufficient to ensure a self-sustaining (autonomous) flame.For flammable mixtures within the flammability range, explosion initiation occurs when the energy transferred from the ignition source exceeds a threshold value, equal to the minimum ignition energy (MIE). Measurement of the minimum ignition energy uses as ignition source capacitive electric discharges, which simulate the sparks produced by static electricity discharges. Besides these high voltage electric sparks, low voltage electric sparks produced by the mechanical breaking of a low voltage circuit containing an inductive component ("break" sparks) are used in studies of the intrinsic safety of electrical equipment working in gaseous environments with explosion hazard, according to IEC 60079-20-1 standard [5]. The characterization of the sensitivity of gases towards ignition by break sparks is made by the minimum ignition current (I_{min}), as recommended by IEC 60079-11 standard method [6]. From the minimum ignition currents one can calculate the associated critical ignition energies, H'_{min}, which can be largely different from the minimum ignition energy by high voltage sparks, H_{min}, commonly used in assessing hazards associated with flames of gaseous mixtures [7–9].

Early studies on break spark ignition reported the minimum ignition currents (MIC) for the most reactive alkane-air, ethylene-air and hydrogen-air mixtures and their critical ignition energies using the test cell developed by PTB (Physikalisch-Technische Bundesanstalt)-Germany and recommended by the standard of the International Electrotechnical Commission [10–16]. Measurements on ethylene-air at ambient initial conditions reported minimum ignition currents between

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Nomenclature

a, b, k, m, n	constants
C_p	molar heat capacity (J mol^{-1} K^{-1})
C	capacitance (F)
CIC	critical (minimum) inert concentration (vol.%)
d	distance (m)
H, H'	ignition energy (J)
I	intensity of current (A)
L	inductance (H)
LEL	lower explosion limit (vol.%)
LOC	limiting oxygen concentration (vol.%)
MESG	maximum experimental safe gap (mm)
MIC	minimum ignition current (A)
MIE	minimum ignition energy (J)
p	pressure (N m^{-2})
UEL	upper explosion limit (vol.%)
V	voltage (V)

Greek

α, χ	constants
β	baric coefficient of minimum ignition currents (–)
∂	partial derivative
λ	thermal conductivity (J m^{-1} K^{-1} s^{-1})

Subscripts

min	minimum
mix	referring to mixture
q	quenching
rel	relative
0	referring to the initial state

0.115 and 0.195 A for mixtures with various equivalence ratios [10,12,14,15]. The results of the experimental determination of MIC for methane/hydrogen-air mixtures [16] were also reported and compared with the MIC of pure methane, in order to classify these mixtures into flammability groups. In recent studies of hydrogen-air ignition by break sparks [17,18], the transient flame kernel development was monitored by high spatial and temporal resolution interferometry and the measurements (ignition delays and threshold currents) were compared with simulated data from of a 3-D reactive flow model with detailed chemical kinetics and molecular transport. The authors were concerned with the significant scattering of experimental results, still present when a new designed apparatus and electrical circuit were used. A better reproducibility of measurements could be obtained by a better control of arc geometry and length, and by the homogeneity of Cd electrode surface. The international standards examine the intrinsic safety in connection to another important feature of explosion-proof equipment: the maximum experimental safe gap (MESG), defined as the maximum distance between parallel surfaces that prevents flame on one side of the flame-path formed by these surfaces from igniting all flammable fuel-air mixtures under test on the other side. The MESG is a property related both to the quenching distance and the minimum ignition current [10,12–15]. According to international standards IEC 60079-20 [5] and IEC 60079-11 [6], I_{min} and MESG are used to classify flammable gases and/or vapors into 2 groups as follows:

- I^{st} Group – for explosive atmospheres occurring in underground (mining) activities;
- II^{nd} Group – for explosive atmospheres encountered in the surface industries, except those parts that belong to underground mines. This group is divided in 3 subgroups: IIA, IIB, IIC. This classification of gases is important for choosing the type of tests which have to be performed in test explosive mixtures using laboratory closed vessels for the electrical/non-electrical equipment, "Ex"-labeled, explosion protected. The test fuels chosen to verify the ignition/nonignition of explosive atmospheres depend on the selected group: for the first group, methane-air mixture is used, for IIA group, propane – air mixture, for IIB group, ethylene –air mixture and for the IIC group, hydrogen – air mixture [14,19,20].

A related work examined recently the intrinsic safety using break sparks produced by a low voltage capacitive circuit, raising similar problems [21].

In the present work, the minimum ignition currents, critical ignition energies and maximum experimental safe gaps for stoichiometric ethylene-air flames diluted with Ar, N_2 or CO_2 at various initial pressures between 20 and 110 kPa and various additive amounts between 8 and 32% (volume percent concentrations) are reported. The critical ignition energies and the maximum experimental safe gaps are calculated from the measured minimum ignition currents using previously validated correlations [12,14,15].

Ethylene was chosen for the present study according to its sensitivity to electrostatic discharges in comparison with other flammable gases. The assessment of its electrostatic hazard, especially in the presence of inert additives, is requested as ethylene is a very reactive fuel and a key intermediate in the oxidation of higher alkanes and alkenes [22]. Measurements on its flames with air will support the understanding and prediction of ignition, propagation and quenching processes of most fuel-air flames. The quenching distances and minimum ignition energies of initiation by high voltage electric sparks of ethylene-air and ethylene-air inert mixtures are available [7,8,23,24] at various initial pressures, ambient and sub-atmospheric. Ignition by low voltage sparks of ethylene-air of various equivalence ratios was examined at ambient initial pressure [10,12,14,15], but no studies on ethylene-air ignition in the presence of inert additives were reported. The present data offer important information in addition to previous works reporting inert gas and initial pressure influence on critical ignition parameters [23–28].

2. Experimental

The experimental set-up contains a vacuum pump, the vacuum line for preparing and evacuation of gases, gas bottles containing fuel and additives, a cylinder for mixture storage, air compressor and the explosion vessel equipped with electric circuit for break spark ignition. The vacuum line is tight at pressures from 10 Pa to 150 kPa, and interconnects the vacuum pump, the gas cylinders with fuel, air and additive, the metallic cylinder for mixture storage and the explosion vessel. The experimental set-up was assembled in the Chemical Kinetics Laboratory, Faculty of Chemistry, University of Bucharest.

The experimental apparatus is in compliance with Annex B of IEC 60079-11 standard [6]. The test cell used to measure minimum ignition currents was similar to that developed by PTB (Physikalisch-Technische Bundesanstalt)-Germany [12,13]. The stainless-steel explosion cell, shown in Fig. 1, has a cylindrical form, with a 9.6 cm internal diameter and 12.2 cm height and it is vacuum- and pressure-tight. It was tested under static pressure within the range of 0.1–5000 kPa [8]. The upper lid is made from a transparent plate from organic glass, doubled by a stainless-steel plate provided with two windows. The central window can be used for magnetic stirring of the gas mixture or for video recording of sparks (as shown in the supplementary material). The eccentric window is used to observe the ignition process either visually or by means of a photodiode. A photodiode S1223 from Hamatsu