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Full Length Article

Additive influence on quenching distances and critical ignition energies of ethylene-air mixtures

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HIGHLIGHTS

- Ignition of ethylene-air-inert (Ar, N_2, CO_2) was studied at variable p_0 (20–110 kPa).
- Stoichiometric mixtures with inert mole fractions within 0.04–0.50 were examined.
- The quenching distances were measured by flanged electrodes technique.
- The pressure dependence of quenching distances, d_q, was described by a power function.
- Measured d_q were used to evaluate critical ignition energies using a correlation model.

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ABSTRACT

The quenching distances of the stoichiometric ethylene-air mixture diluted with argon, nitrogen or carbon dioxide of variable mole fractions within 0.04 and 0.50 were measured at various initial pressures between 20 and 110 kPa using inductive-capacitive sparks produced between flanged electrodes. The pressure dependence of the quenching distance was described by a power function and the corresponding baric coefficients were evaluated by non-linear regression analysis. The experimental quenching distances were compared with those calculated from a model based on the assumption that the critical size of the flame is attained when the rate of heat production equals the rate of heat loss associated with the expansion work during incipient flame growth. The necessary data were taken from literature. The quenching distances d_q measured for various compositions and initial pressures of the flammable mixtures were also used to evaluate the threshold or critical ignition energies H_{min} based on a correlation of the form $H_{min} = k \cdot p_0 \cdot d_q^3$, where p_0 is the initial pressure and $k = 0.445$ is a proportionality constant. The evaluated critical ignition energies matched well the literature data for hydrocarbons, determined by capacitive sparks. The overall kinetic parameters of ethylene combustion (reaction order and activation energy) were determined from the pressure or average flame temperature dependence of quenching distance. The quenching distances, critical ignition energies and overall kinetic parameters were discussed in connection with the amount and nature of added inert.

1. Introduction

The study of flame initiation and quenching is important for risk assessment of accidental explosions of fuel-oxidizer gaseous mixtures and for establishing adequate safety recommendations. Applied research on this topic stems from the present requirements for IC (internal combustion) engines: a better control of fuel efficiency, pollutant formation and engine operation conditions [1]. Other aspects are important, too: the flame stabilization (e.g. in turbojet systems operating at low pressures) and the behavior of flames near cold walls [2,3]. A better understanding of processes occurring in the ignition stage of explosions, such as the contribution of heat and mass transfer processes to formation of the flame kernel, helps predicting the flame extinction in various conditions [4,5].

Being sources of known and reproducible size and energy, the electric sparks were frequently used for flame initiation. The topic is closely bound to the interest for improving combustion in spark ignition engines but may also provide fundamental results able to illustrate and quantify the basic chemical and physical processes through which ignition is produced.

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Nomenclature

a, A_0, A_1, A_2, A_3, b, B, C, k	constants
C_p	heat capacity (J mol^{-1} K^{-1})
d	distance (m)
E	expansion coefficient (–); error (–); energy (J)
H	enthalpy (J); critical ignition energy (J)
i	intensity of current (A)
m	baric coefficient of the quenching distance (–)
n	overall reaction order (–); mole number (mol)
p	pressure (N m^{-2})
q	heat amount (J)
r	radius (m)
R	universal gas constant (8.31 J mol^{-1} K^{-1})
S	burning velocity (cm s^{-1})
t	time (s)
T	temperature (K)
V	volume (m^3)
X	mole fraction (–)

Greek

α	baric coefficient of the laminar burning velocity (–)
∂	partial derivative
ν_l	stoichiometric coefficient of the limiting component (–)
η	flame front width (m)
λ	thermal conductivity (J m^{-1} K^{-1} s^{-1})
ρ_0	density (kg m^{-3})

Subscripts

a	referring to the activation energy
b	referring to the burned gas
c	critical
calc	calculated
chem	referring to a chemical process
e	end
f	flame
i	initial
l	limiting component
meas	measured
min	minimum
q	quenching
tr	transferred
u	referring to the unburned gas
0	referring to the initial state

Several important properties characteristic for flame initiation by electric sparks are the quenching distances d_q, minimum ignition energies (for high-voltage electric sparks) $H_{\min}$ and minimum ignition currents (for low-voltage, or “break” sparks) i_{min} [6,7]. In this last case one can also calculate the associated minimum ignition energies, $H'_{\min}$, but they can be largely different from $H_{\min}$ which is commonly used in assessing hazards associated with flames of gaseous mixtures [8].

The quenching distance is defined as the minimum space between two flat surfaces through which a flame can propagate. The quenching distance is usually measured by the parallel plate method, as the shortest distance between the two electrodes flanged with parallel insulating plates, allowing the deflagration to start and propagate independently [6,9–13,4,14]. Other related methods are based on observation of flame propagation through a diverging tube of circular or rectangular section and measurement of the minimum tube section below which flame cannot propagate [15–18], or measurement of the minimum tube diameter or rectangular slot width that allows a flame to flash back [19,20]; in these two cases, a quenching diameter is obtained.

When an electric spark occurs in a flammable mixture, the ignition takes place if the spark transfers more energy than $H_{\min}$, the minimum ignition energy specific to that mixture. This energy amount should be sufficient to create a hot kernel able to ensure the formation of an autonomous, self-propagating flame [6,21]. In this way, the concept of minimum (critical) ignition energy is bound to the concept of quenching distance, i.e. the critical dimension of the hot kernel that has to be exceeded in order to observe the ignition. According to Lewis and von Elbe [6], the minimum ignition energy is the energy amount supplied by the spark to heat the unburned gas from the spark gap to the adiabatic flame temperature, for reaching the size of “a minimum flame”. In the classical thermal-diffusion theory [6,22] the quenching distance was related also to the flame thickness. Zeldovich [22] proposed that the critical length controlling the initiation of a spherical flame is the flame thickness such that $H_{\min}$ was proportional to the cube of the flame thickness. Later, more accurate models of flame ignition included the effect of preferential diffusion (i.e. the Lewis number effect) and $H_{\min}$ was considered to be proportional to the cube of the flame ball radius [3,6,23,24].

Many empirical correlations between $H_{\min}$ and d_q of the form $H_{\min} = a \cdot d_q^b$ with $2 < b < 3$ have been reported in literature [26], proving their interconnection, explicitly detailed by several theoretical models. The simplest model was proposed by Litchfield [50] assuming that the critical ignition energy is given by the mechanical work $p_0 \cdot V$ required to displace a volume V of gaseous mixture against the pressure p_0, where V is the spherical volume of radius $r = d_q/2$:

$$H_{\min} = p_0 \cdot V = p_0 \cdot (4\pi/3) \cdot (d_q/2)^3 = 0.524 \cdot p_0 \cdot d_q^3 \tag{1}$$

A similar model, which took also into account the initial volume occupied by the gas V^*, was derived by Oancea et al. [25] assuming that the critical ignition energy supplied by a high voltage electric spark equals the expansion work of gases from V^* to the minimal flame with a diameter equal to the quenching distance, $H_{\min} = p_0 \cdot (V - V^*)$. The result of this model was:

$$H_{\min} = k \cdot p_0 \cdot d_q^3 \tag{2}$$

with $k = 0.445$, with $H_{\min}$ given in J and p_0 in kPa.

The correlation was verified by examining of a large number of literature data containing the minimum ignition energies measured in ignitions with capacitive sparks and the quenching distances [6,21,27,28]. Eq. (2) was used to obtain the critical ignition energies of propane-air [29], LPG-air [30], n-butane-air [31,32] and propylene-air mixtures [8,13,26] by using their quenching distances, found in very good agreement with acknowledged direct measurements.

It must be specified that in the pertinent literature related to spark ignition, the term “minimum ignition energy” referred initially [6] to the threshold or critical ignition energy of any fuel-air mixture with composition within the flammability limits. This property exhibits a quasi-parabolic variation with fuel concentration, with a minimum defining the most ignitable mixture. This lowest value, characteristic for a certain fuel is frequently used in explosion hazards evaluation and is also designated as minimum