CARBON DIOXIDE AS INHIBITOR FOR IGNITION AND FLAME PROPAGATION OF PROPANE-AIR MIXTURES

V. Munteanu [*], **D. Oancea** [*] **and Domnina Răzuş** [**]

The effect of CO_2 addition on stoichiometric propane-air mixtures, at ambient pressure and temperature, was analysed for both ignition and flame propagation. The quenching distance significantly increases and the normal burning velocity significantly decreases with the increase of CO_2 content. The ignition and the propagation are differentiated by their activation energies, evaluated from the flame temperature dependence of the quenching distance and the normal burning velocity, respectively.

1. Introduction

An important aspect when referring to prevention of explosions in hazardous areas is the dilution effect of inert gases. If the terms in use are efficiency and cost of operation, one of the common inert gases is carbon dioxide (CO_2).

The influence of CO_2 on several flammability parameters of stoichiometric propane-air mixture has been examined. The inert concentration in the gas phase was scanned within the range of 2%÷32%.

2. Experimental Apparatus & Procedure

The quenching distance, d_q, and the normal burning velocity, S_u, were the two explosion parameters that have been experimentally determined.

The experimental equipment contains three major parts:

– gas control system that covers all gas operations and also monitors the gas flow,

– explosion cell, made of stainless steel, equipped with a spark generator – two electrodes plugged into an electrical discharge circuit, and

– computer-assisted data acquisition system.

The explosion cell, a cylindrical vessel of 6 cm height and 6 cm diameter, made from stainless steel, was fitted with flanged electrodes for quenching distance measurements [1], and an ionisation gauge. The spacing of the electrode tips was varied with a micrometer screw and the spark gap was positioned in the centre of the vessel. The ionisation gauge was placed at a small distance, r_{gauge}, from the spark gap, monitoring the flame propagation

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[*] Department of Physical Chemistry, University of Bucharest, 4-12 Regina Elisabeta Blvd., 70346 Bucharest, ROMANIA

[**] "I.G.Murgulescu" Institute of Physical Chemistry, 202A Spl. Independentei, Bucharest, ROMANIA

in the early stage of explosion, when the process is still isobaric. The space velocity of the flame was calculated with the relationship:

$$S_s = \frac{dr}{dt} \cong \frac{r_{gauge}}{t_{flame}} \tag{1}$$

where t_{flame} is the time from the explosion initiation to the maximum of the ionisation gauge signal, and the normal burning velocity according to:

$$S_u = \frac{1}{E_0} \cdot \frac{dr}{dt} \tag{2}$$

where E_0 represents the expansion factor, and it has been calculated for each test mixture from the thermodynamic properties [2]. The measurements were made at initial ambient pressure and temperature.

3. Results & Discussion

The experimental data are presented in Table 1.

Table 1. Quenching distances, d_q, and normal burning velocities, S_u, for the stoichiometric propane-air explosive mixture, in the presence of CO_2, at ambient initial pressure and temperature

Nr. crt.	c_{CO_2}, %	d_q. mm	E_0	t_{flame}. s	S_s, cm×s^{-1}	S_u, cm×s^{-1}
1	0	1.84	8.00	0.007	294.52	36.81
2	2	1.94	7.88	–	–	–
3	4	2.17	7.73	0.016	138.71	17.95
4	8	2.77	7.42	0.043	49.80	6.71
5	12	3.39	7.10	0.114	18.87	2.66
5	16	4.23	6.86	0.137	15.73	2.29
6	20	5.40	6.56	0.258	8.33	1.27
7	32	non-ignition				

One can observe that CO_2 significantly affects both quenching distances and normal burning velocities of the stoichiometric propane-air mixture; moreover, a 32% of added CO_2 results in reaching the non-ignition domain for propane-air mixture.

The apparent activation energy of the process can be evaluated from the flame temperature dependence of the quenching distance:

$$d_q \sim f_1(T_{flame}) \cdot e^{\frac{E_a}{2RT_{flame}}} \tag{3}$$

where E_a represents the apparent activation energy, in $J \times mol^{-1}$, R is the gas law constant, in $J \times mol^{-1} \times K^{-1}$, T_{flame} is the adiabatic flame temperature, in K, and $f_1(T_{flame})$ is an intricate function of the T_{flame}. The adiabatic flame temperature was calculated from the thermodynamic data [2]. The relation (3) was for the first time proposed [3] knowing the influence of inert dilution on the adiabatic flame temperature [4], and observing that the temperature-dependent pre-exponential factor is less sensitive to temperature variation than the exponential one, thus allowing the evaluation of the apparent activation energy from the effect of "air-dilution" on the adiabatic flame temperature, in the range of lean to stoichiometric concentration of fuel in air. One may go further and apply a correction to the adiabatic flame temperature, by replacing it with the average temperature within the reaction zone [4]:

$$T_{av} = T_0 + 0.74 \cdot (T_{flame} - T_0) \tag{4}$$

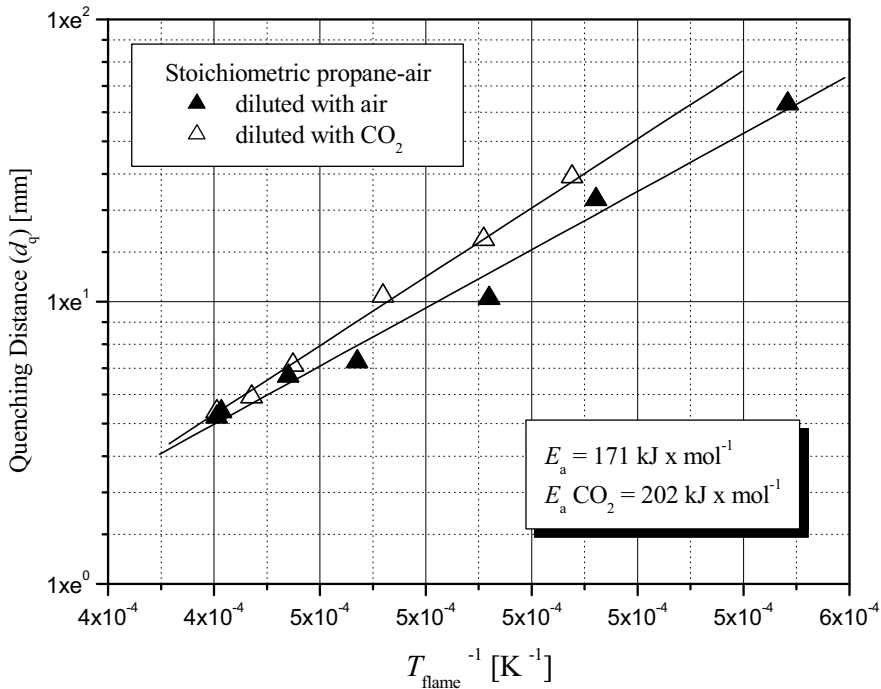T_0 being the initial temperature, in K.

The results are presented in Fig. 1.

Fig. 1: *The influence of* CO_2 *on the apparent activation energy, for stoichiometric propane-air mixture;*
initial pressure 101.3 kPa; initial temperature 298 K.

E_a increases in the presence of CO_2, but the difference is rather insignificant taking into account the experimental errors, and comparing the results with the reported data of Chang

[4], who obtained 196 kJ×mol^{-1} from the dependence of the induction period on the flame temperature, in Vycor tubes.

If one replaces the adiabatic flame temperature with the corrected average temperature, then E_a is 29 kJ×mol^{-1} lowered, maintaining the same difference between propane-air and propane-air diluted with CO_2.

The situation is different when investigating the CO_2 effect on the normal burning velocity.

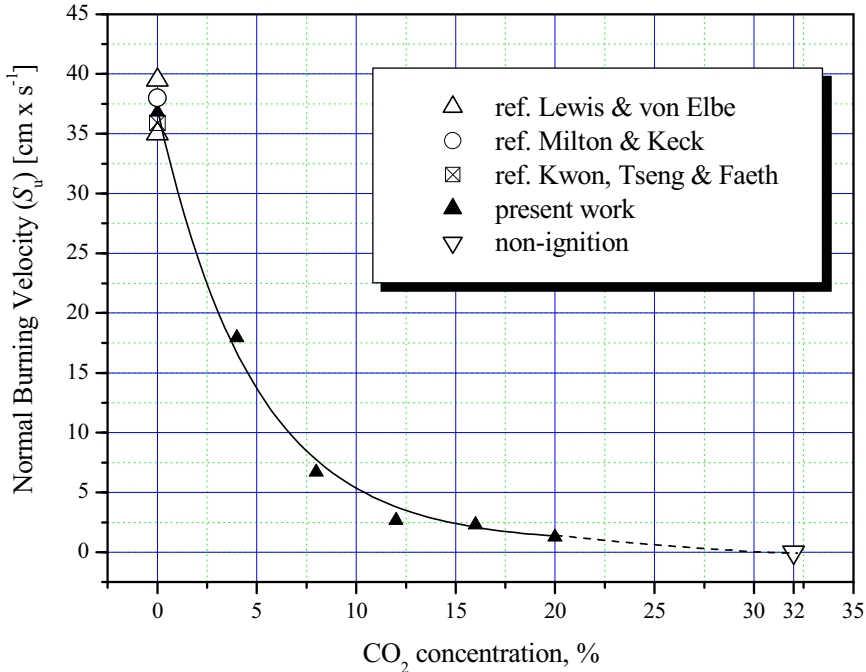Fig. 2 shows the dependence of S_u on the concentration of added CO_2.

Fig. 2: *The effect of* CO_2 *on normal burning velocity, S_u , for stoichiometric propane-air mixture;* initial pressure 101.3 kPA; initial temperature 298 K.

The first observation is that one obtains a very good concordance between the values reported in literature [6÷8] and that obtained in this work, for the stoichiometric propane-air explosive mixture. This gives reasons to rely on the S_u evaluated by such a simple method.

The CO_2 significantly affects S_u, which shows an exponential decrease with the increase of CO_2 concentration, until it reaches a level below 5 cm×s^{-1} for CO_2 concentration larger than 16%.

Similar to the temperature dependence of the quenching distance, the apparent activation energy of the flame propagation can be evaluated from the adiabatic flame temperature dependence of the normal burning velocity [4]:

$$S_u \sim f_2(T_{flame}) \cdot e^{-\frac{E_a}{2RT_{flame}}} \tag{5}$$

bearing the same significance with that of eqn (3).

The same calculated flame temperatures were used to evaluate the apparent activation energy, and the data are plotted in Fig. 3.
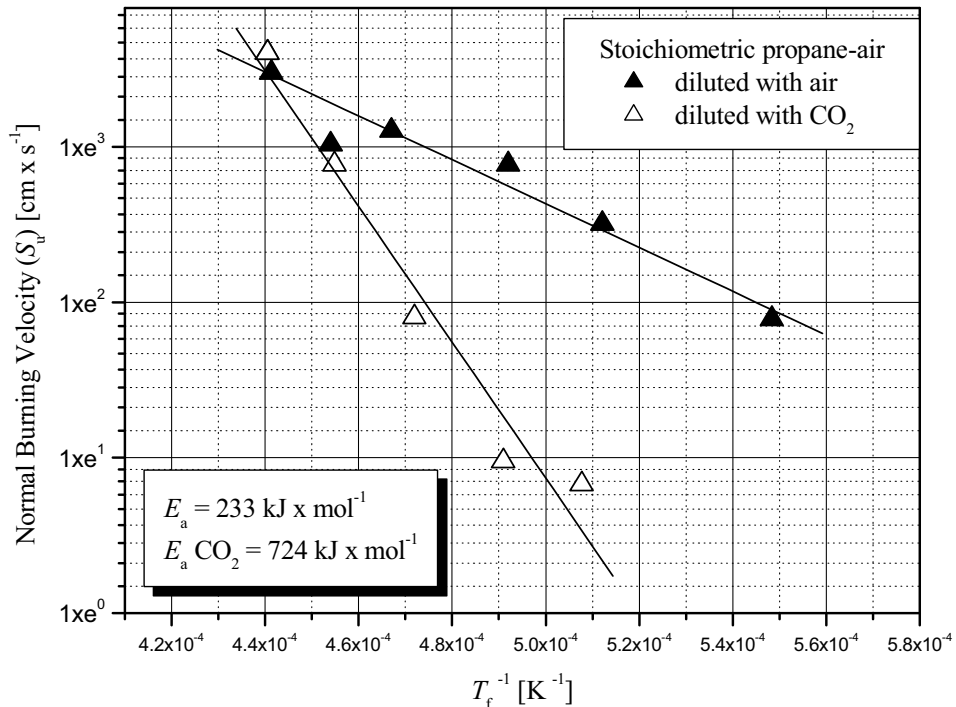

Fig. 3: *Apparent activation energies evaluated from the temperature dependence of the normal burning velocity.*

The addition of CO_2 results in a three times higher apparent activation energy, if either adiabatic flame temperature or average temperature in the reaction zone (590 kJ×mol^{-1} vs. 191 kJ×mol^{-1}) is used.

Analysing the primary results and those obtained from the adiabatic flame temperature dependence of the quenching distance and the normal burning velocity, respectively, it results:

– the addition of CO_2 to propane-air mixture significantly influences both the quenching distance and the normal burning velocity, their differentiation appearing only with respect to their flame temperature dependencies;

– two different behaviours can be distinguished, regarding the influence of inert dilution on the above-mentioned parameters, and these are a very strong effect on the normal burning velocity and a rather insignificant one on the quenching distance;

– the effect of CO_2 on the normal burning velocity is highlighted by the tripling of the apparent activation energy, evaluated from the linear dependence

$$\ln S_u \;=\; f\!\left(\frac{1}{T_{flame}}\right);$$

– the apparent activation energy, evaluated from the linear dependence

$$\ln d_q \;=\; f\!\left(\frac{1}{T_{flame}}\right)$$ does not result in significant change between CO_2-free and

CO_2-added mixtures.

Conclusions

The overall deflagration process can be divided into two major steps: the initiation step, and the propagation step. The quenching distance features the initiation step, and the normal burning velocity the propagation step. One can go further and assume that each step bears its own activation energy.

Carbon dioxide is more than a simple diluting inert; it acts differently on the initiation of the deflagration and on the propagation of the flame.

From the stepping-process analysis, one can say that carbon dioxide affects (in the sense of inhibiting) the propagation step much greater than it does on the initiation step.

Also its efficiency is remarkable, a 32 % of added CO_2 being enough to avoid initiation of the explosion, even with repetition sparks.

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