

Detonability of THDCPD-exo-air mixtures

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Abstract In the frame of industrial risk and propulsive application, the detonability study of JP10-air mixtures was performed. The simulation and measurements of detonation parameters were performed for THDCPD-exo/air mixtures at various initial pressure ($1 \text{ bar} < P_0 < 3 \text{ bar}$) and equivalence ratio ($0.8 < \Phi < 1.6$) in a heated tube ($T_0 \sim 375 \text{ K}$). Numerical simulations of the detonation were performed with the STANJAN code and a detailed kinetic scheme of the combustion of THDCPD. The experimental study deals with the measurements of detonation velocity and cell size λ . The measured velocity is in a good agreement with the calculated theoretical values. The cell size measurements show a minimum value for $\Phi \sim 1.2$ at every level of initial pressure studied and the calculated induction length L_i corresponds to cell size value with a coefficient $k = \lambda/L_i = 24$ at $P_0 = 1 \text{ bar}$. Based on the comparison between the results obtained during this study and those available in the literature on the critical initiation energy E_c , critical tube diameter d_c and deflagration to detonation transition length L_{DDT} , we can conclude that the detonability of THDCPD-air mixtures corresponds to that of hydrocarbon-air mixtures.

Keywords Detonation · Cellular structure · THDCPD-exo · Detonability · Critical energy

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List of symbols

Parameters:

d	tube inner diameter
D	measured detonation velocity
D_{CJ}	Chapman–Jouguet detonation velocity
DDT	deflagration to detonation transition
E_c	critical initiation energy of a spherical detonation
L_i	induction length
L_{DDT}	DDT Length
P	pressure
T	temperature
THDCPD	tetrahydrodicyclopentadiene
u	particule velocity

Subscripts

CJ	Chapman–Jouguet state
ZND	Zel’dovich–von Neuman–Döring state
0	initial state

Greek symbol

λ	detonation cell size
Φ	equivalence ratio
ρ	density
σ	Standard deviation of λ
τ_i	induction time

1 Introduction

The purpose of this work is to study the detonability of gaseous kerosene mixed in various proportions with air in order to evaluate the risk of explosion during transportation in a confined environment. The kerosene investigated

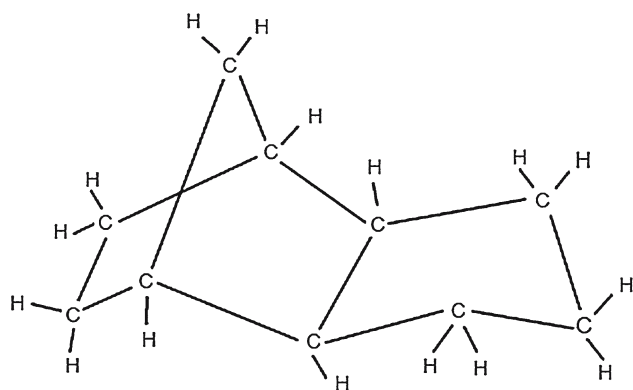


Fig. 1 Structure of THDCPD-exo ($C_{10}H_{16}$)

is tetrahydrodicyclopentadiene (THDCPD; i.e., $C_{10}H_{16}$ also known as JP10) with an exo structure as indicated in Fig. 1. The corresponding C/H ratio and calorific value are 0.625 and 44.5 MJ/kg respectively. Moreover the high energy characteristics of this kerosene are of interest for propulsive application particularly for pulsed detonation engines (PDE). Thus, knowledge of the fuel's detonation characteristics such as the CJ velocity, the CJ pressure and the cell size is extremely valuable.

The detonation properties of THDCPD-air are available in the literature [2, 6, 26]. Austin and Shepherd [2] reported the evolution law of vapor pressure with temperature of THDCPD-exo. They also provided data on detonation wave velocity for stoichiometric conditions and cell size variation as a function of the equivalence ratio, ranging from 0.7 to 1.4, at $P_0 = 1$ bar, as well as a function of pressure, ranging from 0.635 to 1.3 bar at $\Phi = 1$ for THDCPD-exo/air mixtures at $T_0 = 373$ K. Their results exhibited a minimum value of $\lambda \sim 50$ mm at $\Phi = 1.1$. They concluded that the THDCPD-air mixture has the same behavior in terms of cell size measurements as propane-air. Zhang et al. [26] reported cell size measurements for JP-10 at different pressures and temperatures. They showed that T_0 has a minor influence on cell size. A regression law $\lambda = A \cdot P_0^n$ curve was provided for stoichiometric THDCPD-air mixtures based on the values of Ref. [2, 6, 26]. The law shows that $n \sim 1.26$, which corresponds to classical values found for hydrocarbon-air mixtures. Card et al. [6] provided values of the deflagration to detonation transition (DDT) length for lean and rich limits as well as the flame acceleration in a THDCPD-air mixture at 473 K and 2 atm for different equivalence ratios. This allows to measure the length of DDT (L_{DDT}) for $\Phi = 1, 0.8$ and 0.74, these data are reported in Table 1.

In order to compare the relative detonation sensitivity of THDCPD-air mixtures with other compounds, one can consider various detonation dynamic parameters such as: (i) the critical energy E_c to initiate a spherical detonation, (ii) critical diameter d_c of detonation transmission from a tube

Table 1 Length of DDT measured on flame acceleration in obstacle laden tube (results derived from [6]), detonation cell size and normalized parameters

Φ	L_{DDT} (mm)	$\lambda = 19 \cdot L_i$ (mm)	L_{DDT}/λ	d/λ
1.00	1755	27.4	64	2.7
0.80	1936	60.7	31.9	1.2
0.74	2313	91.3	25.4	0.8

to open space, (iii) the detonation propagation limit in a tube and (iv) the deflagration to detonation transition length L_{DDT} . These values can be calculated from a knowledge of the cellular structure width λ . Zel'dovich et al. [25] found that E_c was proportional to the Chapman-Jouguet pressure P_{CJ} and to the induction length L_i . From Desbordes et al. [8], one can express E_c in the form (with $B \sim 425$ for hydrocarbon-air mixtures):

$$E_c \cong A \cdot P_{CJ} \cdot L_i^3 \approx B \cdot \rho_0 \cdot D_{CJ}^2 \cdot \lambda^3. \quad (1)$$

The criterion for successful transmission of a self-sustained detonation from a tube of inner diameter d_c to an open space was first expressed by Zel'dovich et al. [25]. It is proportional to the chemical thickness Δ for 5 different reactive mixtures ($d_c \approx 15\Delta$). The above criterion can be formulated as:

$$d_c = 13 \lambda. \quad (2)$$

Equation (2) applies to most common hydrocarbons/ O_2 / N_2 mixtures in cylindrical tubes [15, 16]. This relationship was further used by Bauer et al. [3] for compositions of hydrocarbons/ O_2 / N_2 at elevated pressures, namely up to 5 MPa.

The detonation propagation limit depends on the roughness of the tube wall. In smooth bore, cylindrical tubes, the limit propagation proceeds through a spinning detonation [14]. This propagation mode corresponds to the coupling between the pulsation of a simple detonation triple point and the acoustic resonance of the tube. The propagation limit can be expressed as:

$$\pi d = \lambda. \quad (3)$$

For obstacle-laden tube (e.g., perforated plate), the detonation propagation limit depends on the comparison between the cell size and the obstacle inside diameter: $d \approx \lambda$ [17].

The DDT length can be used as a criterion for the detonability of the mixture. For $C_3H_8/O_2/N_2$ mixtures the results provided by Pinard et al. [18] obtained in an obstacle-laden tube clearly indicate that L_{DDT}/λ increases with d/λ (cf. Fig. 8). Sorin [20, 21] showed that, given d/λ , the ratio L_{DDT}/λ is

roughly constant depending weakly on the reduced activation energy of the mixture. The above results yielded an estimation of the length necessary for the onset of detonation from flame self-acceleration of the mixture:

$$L_{DDT} \approx 40 \lambda \quad \text{for } d/\lambda \sim 2. \quad (4)$$

We studied the detonation of gaseous THDCPD-exo/air mixture as a function of equivalence ratio ($0.8 < \Phi < 1.6$) and initial pressure ($1 \text{ bar} < P_0 < 3 \text{ bar}$) at an initial temperature $T_0 \sim 373 \text{ K}$. In the first place, the detonation properties were calculated using a thermochemical code, STANJAN. The detonation velocity and the cell width λ were then measured. Based on the comparison of the calculated critical initiation energy E_c or DDT length with the literature data concerning common hydrocarbons-air mixtures, the detonability of the THDCPD-air mixture is discussed.

2 Experimental details

We studied the detonation of THDCPD-exo and air mixtures for various equivalence ratios and initial pressures. The THDCPD-exo used contains a mass fraction of 99% of $C_{10}H_{16}$. Since THDCPD-exo is liquid at ambient pressure and temperature, the experiments must be performed at elevated temperature. We checked the evolution of THDCPD-exo vapor pressure as a function of temperature and confirmed the data given by Austin and Shepherd [2] for $323 \text{ K} < T_0 < 373 \text{ K}$. In order to detonate a gaseous mixture at $0.8 \leq \Phi \leq 1.6$ and $1 \text{ bar} \leq P_0 \leq 3 \text{ bar}$ we need a partial pressure in the range of 15 to 80 mbar; i.e., maintain a temperature of 335 to 375 K. Therefore, we studied the detonation of gaseous mixtures for $365 \text{ K} < T_0 < 385 \text{ K}$ (cf. Table 2) in order to make sure that there is no cold point in the tube and that all the THDCPD-exo is vaporized.

The experiments were conducted in a heated 5 m steel tube with an inner diameter $d = 52 \text{ mm}$. The reactants is injected in the heated tube on the basis of its partial pressure. A recirculation line is installed and allows the mixing of the reactants. This mixing procedure arbitrarily takes place for at least 15 min in order to ensure the homogeneity of the mixture. The detonation tube is immersed in an oil bath which is externally heated and conveyed into a recirculation line (see Fig. 2). The temperature is measured by means of two thermocouples positioned in the oil bath along the tube. The value of T_0 indicated in Table 2 is an average. The detonation is ignited by means of a pyrotechnic device (i.e. a mercury fulminate inflammer coupled with a detonator HERICA n°8) at one end of the tube.

The detonation velocity and pressure were recorded by means of two pressure transducers (spaced by 760 mm) which

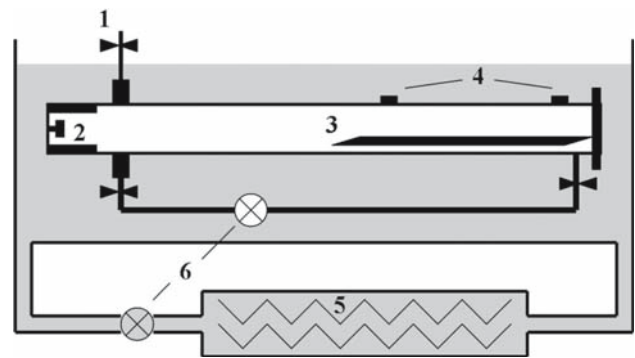


Fig. 2 Scheme of the experimental device; 1 injection point, 2 pyrotechnic initiation, 3 smoked foil, 4 pressure transducers, 5 electrical heating, 6 recirculation pump

Table 2 Summary of experimental results

P_0 (mbar)	T_0 (K)	Φ	λ (mm)	σ (mm)	D (m/s)	D_{CJ} (m/s)
2997	375	0.81	35.8	9.4	1691	1726
3018	379	0.92	21.8	5.1	1746	1771
1974	379	0.91	33.5 ^a	7.1 ^a	1814	1764
3013	368	0.99	17.2	3.0	1746	1803
2015	372	1.01	27.4	4.4	1777	1796
1501	377	0.99	39.5	8.6	1767	1791
2971	377	1.21	13.0	2.3	1823	1839
2023	375	1.18	21.0	3.5	1826	1832
1512	371	1.17	30.2	8.9	1788	1827
1015	384	1.21	48.4	8.5	1737	1821
2907	379	1.40	15.1	1.8	1831	1840
1991	381	1.38	23.8	4.2	1830	1836
1520	365	1.44	32.4	5.3	1811	1833
2956	386	1.58	21.1	4.5	1823	1820
2023	375	1.62	32.7	5.3	1826	1818

^a Corrected value due to measured overdriven velocity

are located at 4 m from the ignition end. We used KISTLER 603B pressure transducers which have a frequency response of $1 \mu\text{s}$ and pressure signals are recorded using a TEKTRONIX oscilloscope. Since the transducer is 5 mm diameter and the detonation remains below 2,000 m/s, the sampling rate was chosen at 10^6 samples per second to accurately capture the CJ peak. This system induces an error of the order of 2% on the velocity measurement, indicated by the error bars on Fig. 3. A 1.5 m long 55 mm wide smoked foil was inserted in the tube in order to record the detonation structure. The detonation cell size λ is the average of at least 20 measured structures along the foil (the standard error on the calculation is reported in the Table 2 and by error bars shown on Figs. 5 and 6).

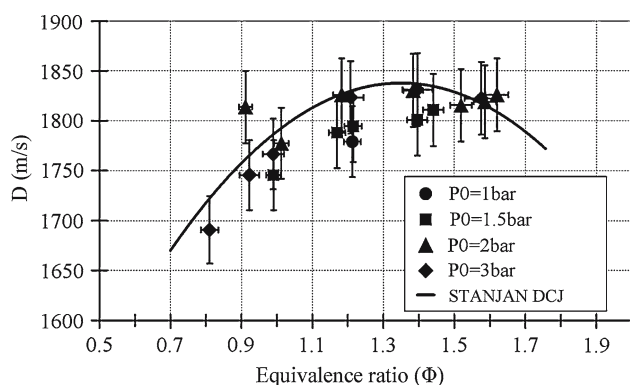


Fig. 3 Comparison between experimental and theoretical detonation velocity; error bars correspond to the characteristic error of velocity measurement

3 Results

3.1 Numerical results

We performed the calculation of detonation thermochemical parameters using the STANJAN code [19]. This code allows the three characteristic parameters of the CJ detonation to be calculated, namely, the CJ velocity D_{CJ} , the normalized pressure P_{CJ}/P_0 and the induction time and length, τ_i and L_i . The STANJAN code uses the CHEMKIN-II chemical kinetics package [12] to calculate the chemical composition variation and thermodynamic parameters (ρ , T) in the reaction zone behind the precursor shock (the ZND state). STANJAN also provides the time that corresponds to the maximum of temperature variation $[dT/dt]_{max}$, i.e. the induction time τ_i .

The detailed reaction mechanism was provided by Williams [13,24]. It involves 55 chemical species and 251 reactions, and is appropriate for the ignition mechanism of THDCPD and heptane mixtures as well as for NO_x formations in addition to the classical reaction mechanism C₁–C₃ [22]. This reaction mechanism is validated for $0.6 < \Phi < 2.0$ and $1 \text{ bar} < P_0 < 9 \text{ bar}$.

Calculated theoretical and experimental velocities, D_{CJ} and D respectively, are shown in Fig. 3. The theoretical calculations exhibit a maximum for $\Phi \sim 1.35$ with $D_{CJ} \sim 1840 \text{ m/s}$ and $P_{CJ}/P_0 \sim 19.4$. These values are consistent with classical hydrocarbon–air mixtures measurements. Theoretical and experimental values show a good agreement with respect to the inherent experimental error (less than 2%). We then checked that the observed detonation was close to the self-sustained propagation and that the detonation was established in the tube.

Since the experimental device did not allow us to perform measurements at $\phi \geq 1.4$ for $P_0 = 1.5 \text{ bar}$, the experimental velocity curve seems to increase monotonically. We clearly see that for different initial pressures (2 and 3 bar), the detonation

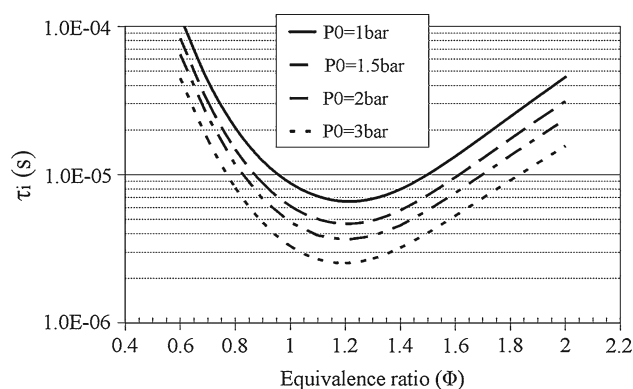


Fig. 4 Evolution of induction time τ_i as a function of equivalence ratio Φ

velocity tends to decrease for $\phi \geq 1.4$ as predicted by the theoretical calculations, which indicates that the trend for $P_0 = 1.5 \text{ bar}$ is due to a lack of measurements. Moreover, we notice that the discrepancy between theoretical and measured value is larger when $P_0 \leq 1.5 \text{ bar}$, i.e., for mixtures with larger detonation cell sizes (see Figs. 5 and 6). This observation can be explained as follows. For mixtures with λ near d , the heat loss at the tube wall modifies the detonation propagation and slows it down.

The computed induction time τ_i is shown in Fig. 4 as a function of equivalence ratio Φ for initial pressures $P_0 = 1, 1.5, 2$ and 3 bar at $T_0 = 373 \text{ K}$. The curves show a minimum value of τ_i at $\Phi \sim 1.2$ regardless of the initial pressure. Since the detonation cell size is proportional to the induction length [23] one should observe a minimum λ for equivalence ratio around 1.2, as confirmed in [2] at $\Phi \sim 1.1$. We checked the variation of τ_i as a function of initial pressure P_0 and found that the evolution is consistent with $\tau_i = A \cdot P_0^{-n}$ keeping a global reaction order $n = 0.89$. This reaction order is lower than the one exhibited by experimental measurements (from the $\lambda = k \cdot P_0^{-n}$ law) of hydrocarbon–air mixtures ($n \sim 1.2$).

3.2 Experimental results

The detonation cell width of THDCPD-exo/air mixtures was measured at $T_0 \sim 375 \text{ K}$ as a function of the equivalence ratio and initial pressure. The data are presented in Table 2. The discrepancy on the initial temperature is less than 3%. A simple result at $\Phi = 0.91$ exhibits a velocity that is higher than the error on the measurement. This discrepancy is certainly due to a late onset of the detonation which remains overdriven. The detonation cell size depends on the local velocity of the detonation through the following relationship [7]:

$$\frac{\lambda}{\lambda_{CJ}} \cong \frac{D}{D_{CJ}} \cdot \exp\left(\frac{E_a}{R \cdot T_{ZND}} \cdot \left[\left(\frac{D_{CJ}}{D}\right)^2 - 1\right]\right). \quad (5)$$

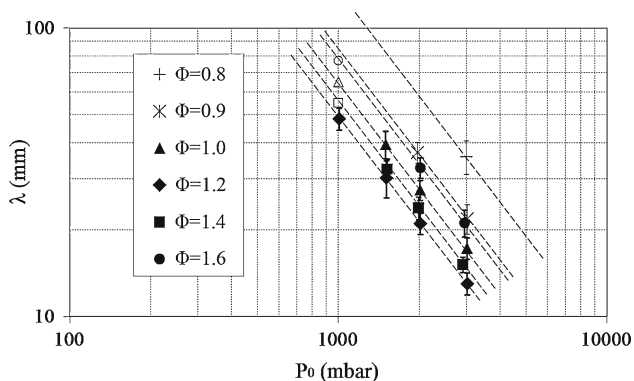


Fig. 5 Variation of detonation cell size λ as a function of initial pressure P_0 for different equivalence ratios Φ in THDCPD-exo/air mixtures at $T_0 \sim 373$ K; *dark symbols*: measured values, *hollow symbols*: estimated values; *dot lines*: estimated regression $\lambda = A \cdot P_0^{-1.2}$; *vertical bars*: standard errors σ from Table 2

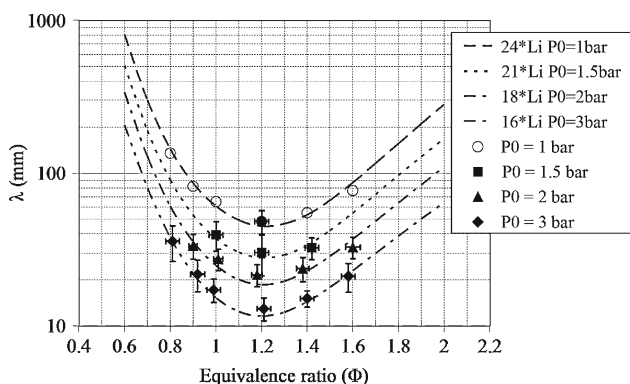


Fig. 6 Variation of detonation cell size λ as a function of equivalence ratio Φ for different initial pressures P_0 in THDCPD-exo/air mixtures at $T_0 \sim 373$ K; *dark symbols*: measured values, *hollow symbols*: estimated values; *dot lines*: estimated values with numerical simulation based on $\lambda = k \cdot L_i$; *vertical bars*: standard errors σ from Table 2

The activation energy for THDCPD-exo mixtures was set equal to 180 kJ/mol [9], and for the data at $\Phi = 0.91$ and $P_0 \sim 2$ bar the ZND temperature (temperature behind the shock front) is $T_{ZND} = 1621$ K which yields $\lambda/\lambda_{CJ} = 0.86$. Although the measured value of λ is 29 ± 6.1 mm, we estimate that the actual value is $\lambda_{CJ} = 33.5 \pm 7.1$ mm.

Figure 5 exhibits the evolution of the detonation cell size λ as a function of initial pressure ($1 \text{ bar} < P_0 < 3 \text{ bar}$) of THDCPD-exo/air mixtures for various equivalence ratios Φ . In the Figs. 5 and 6, the vertical error bars correspond to the standard deviation of the cell size measurement made on the smoked foil. The detonation cell size follows the $\lambda = A \cdot P_0^{-n}$ law within the range of equivalence ratio that was investigated with $n \approx 1.19$. Moreover the data collected in the detonation database seem to indicate that the variation of equivalence ratio has little influence on the parameter n . The data at $P_0 = 1$ bar were estimated so that the regression law

matches the requirement $n = 1.2$. This evolution is common to most hydrocarbon-air mixtures where $1.08 < n < 1.22$.

Figure 6 exhibits the evolution of the detonation cell size λ as a function of the equivalence ratio ($0.8 < \Phi < 1.6$) for different initial pressures P_0 . For each initial pressure the detonation cell size shows a minimum at $\Phi = 1.2$ as predicted by the numerical calculation. The numerical simulations allow to calculate the induction length L_i derived from the relation:

$$L_i = \tau_i \cdot (D_{CJ} - u_{ZND}). \tag{6}$$

As commonly observed, the detonation cell size of hydrocarbon-air mixtures varies proportionally to the induction length L_i [23]. The coefficient of proportionality is $k = \lambda/L_i = 29$ for a large number of mixtures. If we compare the evolution of $k \cdot L_i$ with the equivalence ratio (Φ), we see that for each initial pressure these results are in good agreement with experimental values. The parameter k is merely evaluated in order that the $k \cdot L_i$ curve fit the experimental data. We estimate then that for $P_0 = 1$ bar, k is closed to 24. Since the calculated induction time τ_i varies according to $P_0^{-0.89}$ and the cell size λ as $P_0^{-1.2}$, we observe that k varies with the initial pressure: $k = 24, 21, 18, 16$ respectively for $P_0 = 1, 1.5, 2$ and 3 bar.

4 Discussion

We aim at providing the appropriate data to assess whether a THDCPD-exo/air mixture may raise a concern for industrial application. We can estimate detonation hazards of such mixture by comparing values of the critical initiation energy E_c , critical tube diameter d_c and DDT length L_{DDT} with data available in the literature for such parameters. In order to estimate E_c and d_c we first calculated the theoretical values of λ using the approximation $24 \times L_i$ based on the knowledge of τ_i derived from numerical simulation at $P_0 = 1$ bar. Then E_c and d_c were calculated from Eqs. (1) and (2).

The evolution of critical energy E_c as a function of the equivalence ratio Φ for THDCPD-exo/air mixtures is shown in Fig. 7. Like the cell size evolution, E_c exhibits a minimum value around $\Phi = 1.2$. If we compare E_c values with those of propane and ethane-air mixtures we conclude that the detonability of THDCPD-air mixtures is equivalent to hydrocarbon-air mixtures in terms of equivalence ratio variation. Moreover in order to study the application to propulsion we compared THDCPD-air mixtures with JP4 and *n*-hexane-air mixtures for stoichiometric conditions. The comparison shows that the detonability of these mixtures is similar.

An assessment of detonability of THDCPD-air mixtures based on the definition of the detonation propagation limit

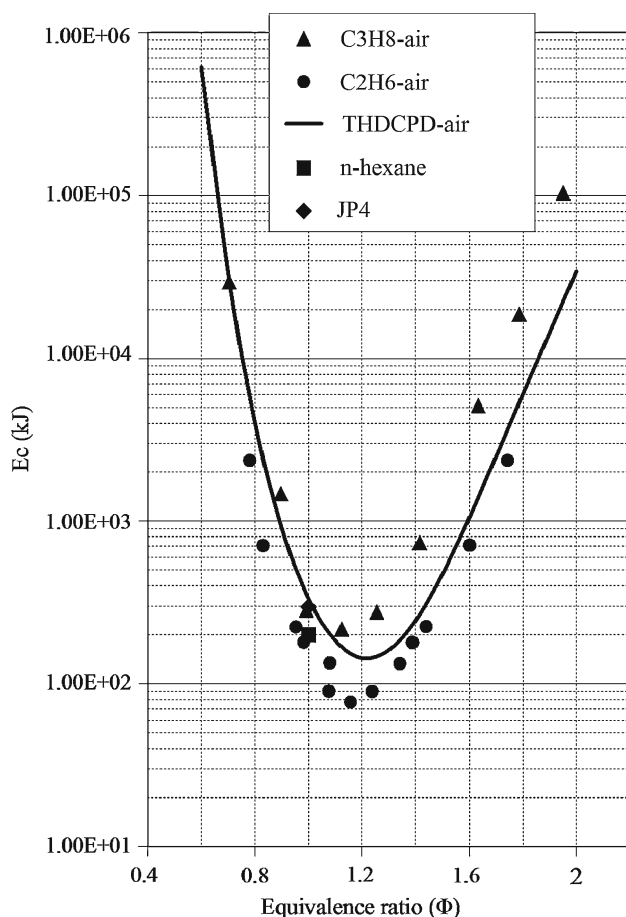


Fig. 7 Variation of critical initiation energy E_c as a function of equivalence ratio Φ for different hydrocarbon mixtures; C_3H_8 -air: [10], C_2H_6 -air: [5], *n*-hexane and JP4: [4]

(Eq. (3)) or critical tube diameter (Eq. (2)) yields two conclusions:

- a self-sustained detonation will not exist in a cylindrical tube of inner diameter lower than 0.014 m
- the self-sustained detonation transmission to open space will not be possible from a cylindrical tube with a diameter less than 0.6 m.

The evolution of L_{DDT}/λ with d/λ for $C_3H_8/O_2/N_2$ mixtures [18] and for THDCPD-exo mixtures at $P_0 = 2$ bar [6], see Table 1, is shown in Fig. 8 (with λ estimated from [11]). The knowledge of the length required for a flame to transit into a detonation is a good way to estimate the relative detonation sensitivity of a mixture [20,21], even if the detonation is not directly initiated but proceeds through a long flame propagation which can be ignited by a low energy deposit. The DDT investigated by Pinard et al. [18] and Card et al. [6] were observed in the same geometrical conditions; i.e., a cylindrical tube of $d = 100$ mm with equidistant perforated plates (blockage ratio of 0.43) located along the tube. The

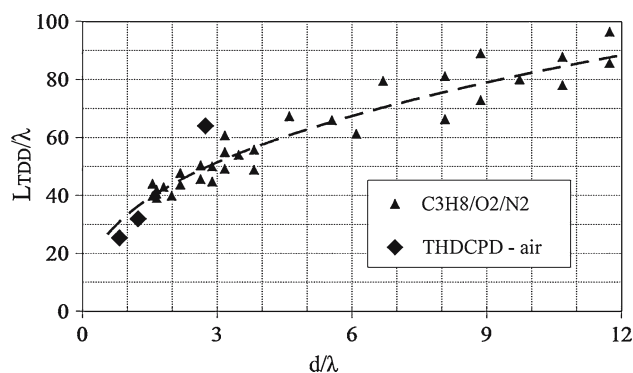


Fig. 8 L_{DDT}/λ versus d/λ ; $C_3H_8/O_2/N_2$: results from [18], THDCPD-air: results from [6], the dot line corresponds to power regression fit for [18] values

geometrical influence was equivalent and had the same effect on the flow. The results may thus be comparable. The data obtained for the THDCPD-air mixture are in agreement with those of propane. Thus in the same geometrical conditions, the DDT behavior of a THDCPD-air or propane/ O_2/N_2 mixture will be similar with respect to the detonation cell size to diameter ratio.

5 Summary and conclusion

The calculation of detonation parameters (i.e., velocity, pressure and induction length) and related measurements were performed for THDCPD-exo/air mixtures at various initial pressures ($1 \text{ bar} < P_0 < 3 \text{ bar}$) and equivalence ratios ($0.8 < \Phi < 1.6$). In order to have sufficient partial vapor pressure of THDCPD in the mixture, the detonation tube was heated up to an initial temperature $T_0 = 375 \pm 10 \text{ K}$.

The numerical simulations of the detonation were performed by means of the STANJAN code and a detailed kinetics of Williams for THDCPD combustion. It shows that the detonation velocity D_{CJ} exhibits a maximum ($\sim 1840 \text{ m/s}$) for $\Phi = 1.35$. Moreover the variation of the induction time (τ_i) as a function of equivalence ratio shows a minimum at $\Phi = 1.2$ regardless of the initial pressure.

The experimental study deals with measurements of the detonation velocity and cell size. The measured velocity is in agreement with the calculated values. The cell size measurements show a minimum value for $\Phi \sim 1.2$ and the calculated induction length L_i is proportional to the value of the cell size with a coefficient $k = 24$ at $P_0 = 1$ bar.

Comparison of the present results with available data in the literature on the detonation critical initiation energy E_c , critical tube diameter d_c and length of deflagration to detonation transition L_{DDT} , allows to conclude that THDCPD-air mixtures have the same level of detonability as propane-air mixtures, which can be regarded as appropriate representative of hydrocarbon-air mixtures.

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