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► **To cite this version:**

Zakaria Bouali, J. Reveillon, Cécile Pera. Development of an extended reactor configuration to analyze preferential segregation impact on spray autoignition. *Fuel*, 2021, 302, pp.120869. 10.1016/j.fuel.2021.120869 . hal-03388334

HAL Id: hal-03388334

<https://ifp.hal.science/hal-03388334v1>

Submitted on 13 Jun 2023

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Development of an extended reactor configuration to analyze preferential segregation impact on spray autoignition

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Abstract

The chemical reactor concept, usually based on a homogeneous mixture, is extended to non-homogeneous two-phase flows to study the impact of preferential segregation on the autoignition of an n-heptane spray in air. To introduce inhomogeneities, two-dimensional reactors are resolved thanks to direct numerical simulations (DNS) with Eulerian/Lagrangian description to follow the evolution of the carrier phase and the dispersed evaporating droplets, respectively. Two-way coupling is considered through exchange of mass, momentum, and energy between the carrier-gas phase and the dispersed phase. A chemistry mechanism with 29 species and 52 reactions was chosen to describe the chemical reaction paths. Several simulations were performed with various initial gas temperatures (i.e. low, intermediate and high) and various geometrical and physical characteristics of the preferential segregation. Results confirmed that evaporative cooling, vapor mass quantity and turbulence mixing play important roles in the two-phase flow autoignition. We discussed the ignition delay as well as the

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Nomenclature

$()^{st}$	Stoichiometric value	S_L	Segregation level
$()_0$	Initial values	T_0	Initial gas temperature
$()_k$	Droplet properties	Y_v	Fuel vapor mass fraction
$()_\alpha$	Properties of α^{th} species	Y_α	Mass fraction of α^{th} species
δ_c	Characteristic cluster size	Z	Mixture fraction
$\overline{\xi^2}$	Density variance	Z_{MR}	Most reactive mixture fraction
$\bar{\xi}$	Mean spray density		
ξ	Local density of spray	0D	Zero-dimensional
a_0	Initial diameter of droplets	2D	Two-dimensional
a_k	droplet diameter	3D	Three-dimensional
$B_{m,k}$	Mass transfer coefficient of k^{th} droplet	DNS	Direct numerical simulation
		HT	High temperature
$B_{T,k}$	Heat transfer coefficient of k^{th} droplet	IT	Intermediate temperature
		LT	Low temperature
NTC	Negative Temperature Coefficient	PDF	Probability density functions

location of the first hot spots according to the initial gas temperatures. The dependence of most reactive mixture fraction on initial carrier-gas temperature is non-monotonic, thus there is a strong correlation between the first autoignition location and low scalar dissipation rate. On the other hand, the autoignition can start whatever the vorticity magnitude is.

Keywords: autoignition, two-phase flows, evaporation, preferential segregation, *NTC*, DNS

1. Introduction

During the last decades, combustion chamber modeling has been extensively developed thanks to better understanding in various physical phenomena interacting in engines from the liquid fuel injection, down to combustion processes and gas exhaust. However, for their environmentally and economically optimization, model accuracy still needs to be improved. Many physical and chemical phenomena interact within the chamber. Pollutant formation directly depends on the combustion which itself depends on the mixing quality and local thermodynamic conditions. After primary atomization close to the injector tip, a secondary atomization generates a dispersed spray. These droplets are small enough to undergo the turbulent velocity fluctuations and they evaporate according to the local flow properties (temperature, pressure, fuel's vapor concentration). Modern direct injection engines now use multiple split-injections that can even overlap each other (a spray can be injected while the previous one is still not completely dispersed or has started to autoignite). From a modeling point of view, it is necessary to correctly represent ignition while mixing and droplet evaporation still occur.

Nowadays, optimization of direct injection car engines have lead to a strong increase of the injection pressure and all engine cycles are so accurately cadenced that they generally overlap each other. Thus, when ignition occurs, mixing and evaporation processes are mostly still in progress. Depending on mixing and evaporation, ignition delay defines the combustion regimes and the structure of the resulting flames.

Previous numerical works on the autoignition in single-phase flows have revealed that ignition occurs first in regions of the flow characterized by: (i) a well-defined value of mixture fraction named "most reactive mixture fraction" and (ii) a low scalar dissipation rate. This result was obtained with two-dimensional

28 (2D) DNS of Homogeneous Isotropic Turbulence (HIT) considering a simplified
29 chemistry model [1]. It was later confirmed by other studies including three-
30 dimensional (3D) effects [2], more complex flow [3] and/or complex chemistry
31 [4]. Viggiano [4] highlighted the importance of taking into account at least a
32 skeletal chemistry mechanism to properly predict the ignition. Using a chem-
33 istry for hydrogen-air, the effect of temperature heterogeneity as well as detailed
34 chemical paths on the autoignition was investigated by [5]. They numerically
35 study the effects of different temperature distributions on the autoignition of a
36 turbulent homogeneous mixture. They found that the presence of a hot core
37 gas leads to an increase in burn duration, while a cold core gas may lead to an
38 undesirable slow combustion of the end gas. It is important to emphasize that
39 this temperature variation topic is omnipresent in the framework of two-phase
40 combustion. However, even if gaseous flow investigations offer major ground
41 knowledge, there are numerous issues that lack answers. And it has been only
42 very recently that research teams were able to tackle some of them. Despite
43 the large number of papers dedicated to single-phase flows [6], considerably less
44 studies were devoted to spray autoignition. These last ones adopt, in general, a
45 coupling of the formalism DNS for carrier phase with the particle-source in cell
46 model for dispersed phase. The autoignition of n-heptane sprays at high tem-
47 perature was investigated numerically by Wang and Rutland [7, 8] considering
48 a 2D configuration and a skeletal mechanism of n-heptane. They found that an
49 increase of the initial gas temperature leads to a decrease of the autoignition
50 delay, while an opposite behavior was observed for the global equivalence ratio.
51 The impact of initial droplet size on the autoignition location was investigated
52 by Schroll et al. [9] using three-dimensional DNS with one-step global chemistry.
53 They performed three simulations with the same initial global equivalence ratio
54 but different initial droplet size. They found that the initial droplet size strongly

55 impacts the autoignition delay but does not affect the value of the most reactive
56 mixture fraction. Thereafter, 3D-DNS with detailed chemistry were performed
57 to investigate the impact of spray properties, the turbulence intensity and shear
58 on the ignition. Borghesi et al. [10] have been analyzed the effect of the initial
59 turbulence in the carrier gas, the global equivalence ratio in the spray region
60 and the initial droplet size distribution of the spray autoignition. Their results
61 revealed that (i) turbulence reduces the ignition delay time as it promotes mix-
62 ing between air and fuel vapor, (ii) a larger number of ignition kernels is directly
63 related to high values of global equivalence ratio, due to higher probability of
64 the most reactive regions, and (iii) spray polydispersity results in the occurrence
65 of ignition over a wider range of mixture fraction values. The impact of shear
66 on evaporation and spray autoignition process was analyzed by Abdelsamie et
67 al. [11]. They found that the autoignition delay time strongly depends on
68 the droplet diameter and the jet velocity, while it is far less sensitive onto the
69 equivalence ratio. They observed also that with high shear, ignition occurs on
70 the lean side, and peak heat release is found near stoichiometry in a premixed
71 combustion mode. In the absence of shear, ignition occurs simultaneously over
72 a broad range of mixture fractions, and peak heat release is found for a rich
73 mixture involving both premixed and non-premixed flames. In spark ignition
74 context, Neophytou et al. [12, 13] attest that the success of ignition was linked
75 to the number of droplets engulfed by the spark. They observed a strong depen-
76 dence of the reactive processes with the spray properties. It appears that the
77 heat release rate is lower than in the equivalent gaseous problem and is limited
78 by the evaporation rate. They also observed a large scatter of heat release and
79 species in mixture fraction space making difficult the development of adapted
80 combustion models.

81 Despite the aforementioned researches, there are numerous issues concerning

82 autoignition in sprays that lack answers, particularly in the low and intermedi-
83 ate temperature range where *cool flame* and Negative Temperature Coefficient
84 (*NTC*, i.e. autoignition delay increases with temperature) phenomena take
85 place.

86 This issue has been studied by 2D-DNS considering detailed chemistry in
87 purely gaseous context. The influence of variations in the initial temperature
88 and the ratio of turbulence to ignition delay timescale on multi-stage ignition
89 of a lean n-heptane/air mixture was studied by Yoo et al. [14]. They observe
90 that, (i) regardless of the mean initial temperature, the mean heat release rate
91 increases more slowly with increasing thermal stratification and (ii) ignition
92 delay decreases with increasing thermal stratification for high mean initial tem-
93 perature relative to the *NTC* regime. It is, however, increased with increasing
94 thermal fluctuations for relatively low mean initial temperature resulting from
95 a longer overall ignition delay of the mixture. More recently, Krisman et al.
96 [15] have been studied the behavior of the low-temperature chemistry and its
97 influences on the high-temperature ignition. They have discussed the location
98 of first-stage autoignition and highlighted its impact on the second-stage au-
99 toignition delay.

100 Conversely, studies dedicated to autoignition at low and intermediate tem-
101 perature in two-phase context was limited to: (i) a non-dimensional homoge-
102 neous reactor [16] or (ii) one isolated droplet one-dimensional configurations
103 [17, 18] and even if there are still unaddressed issues in these basic configura-
104 tions, the dispersing nature of the spray and the preferential segregation effects
105 lead to other difficulties and to combustion regimes that could not be char-
106 acterized. Indeed, as described by Aggarwal [19] from a qualitative point of
107 view, there are three dominant ignition modes and corresponding ignition de-
108 lays: droplet ignition, group ignition and spray ignition. First, similarly to the

109 combustion regimes described by Chiu et al. [20, 21], ignition can occur either
110 individually for droplets separated by a wide interspace, or around clouds of
111 droplets if they are too close to each other to allow for diffusion of heat inside
112 the cloud. A third mode: the spray ignition mode, considers a global ignition of
113 the spray for droplets close to each other but surrounded by a hot atmosphere.
114 In the context of internal combustion engine, where droplets are dispersed in a
115 hot environment, this last mode is of particular interest. Usual homogeneous
116 reactor studies offer better insights of the impacts of a liquid phase onto ig-
117 nition properties but they cannot display any geometrical aspect of the spray
118 dispersion since there is no spatial information. However, in practical systems,
119 preferential segregation of droplets early appears because of strong turbulent
120 mixing [22]. It leads to the formation of non-homogeneous pockets of fuel vapor
121 that cannot be characterized by non-dimensional approaches though there is a
122 direct impact on combustion regimes and model performances.

123 The objective of the present work is to use direct numerical simulation as-
124 sociated with a Lagrangian dispersed phase solver (DNS-DPS) to:

- 125 (i) extend homogeneous reactors studies to multiple clouds or clusters sys-
126 tems, the size and droplet density of which are prescribed. It allows ac-
127 counting for the geometrical aspect of the spray in addition to the funda-
128 mental thermodynamical properties.
- 129 (ii) take into account the effect of preferential segregation of droplets on au-
130 toignition and various ignition shell.

131 The segregation is defined by the spray density variance and the characteristic
132 size of the clouds (or clusters) of droplets. Of course, three-dimensional com-
133 putations would have been better, however, computational's costs would have
134 been skyrocketing to simulate large enough configurations to carry out accurate
135 statistics (statistics need multiple integral length scales performed over long

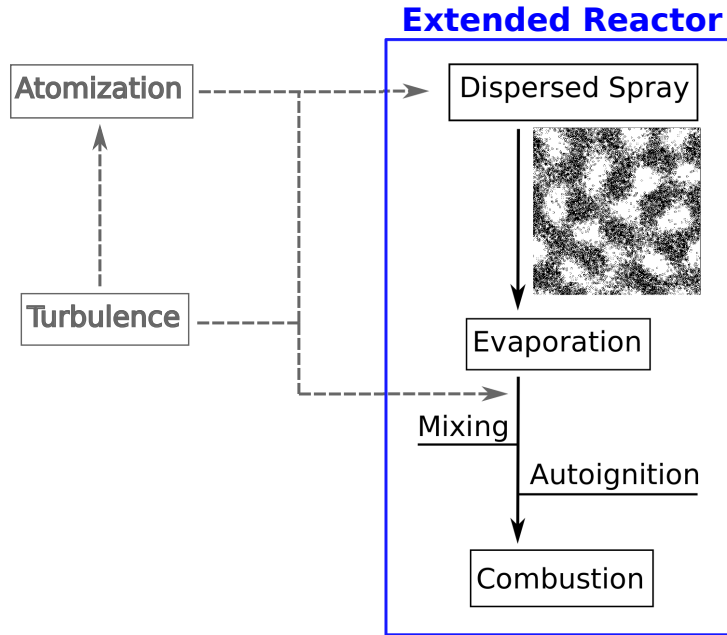


Figure 1: Phenomena taken into account by the Extended Reactor (ER).

136 enough rotating turbulent times). However, as underlined by Wang and Rut-
 137 land [8] and shown by Sreedhara and Lakshmisha [2], the topology of autoigni-
 138 tion spots remains similar enough irrespective of whether the flow is represented
 139 in two or three dimensions. It remains a technical challenge that still need to
 140 be addressed if a wide range of droplet clouds have to be considered. As a
 141 first step, one wishes to separate mixing and turbulence effects from the spray
 142 topology. Therefore, the droplets are first embedded in a quiescent atmosphere
 143 although convection due to gas dilatation is characterized. This ‘Extended Re-
 144 actor’ (ER) configuration must be considered as a 2D (or 3D) extension of a
 145 usual non-dimensional reactor that is not able to consider spray preferential
 146 segregation. Figure 1 summarize the physical phenomena involved or that can
 147 be involved in our ER. This multiple dimension reactor involved, at least, phe-
 148 nomena outlined in blue color and can also integrate the effect of the turbulence
 149 and the atomization.

150 **2. Physical model and configuration**

151 *2.1. Physical model*

152 All simulations have been performed with the DNS code `Asphodele` [23, 24].
153 This reactive solver consider an Eulerian description of the gas coupled to a
154 Lagrangian description of the liquid phase. The coupling between both phases
155 is achieved thanks to the introduction of source terms in the mass, momentum,
156 and energy equations for the continuous gas phase, while the effects of the gas
157 phase are introduced in the form of boundary conditions applied at the boundary
158 layer (gas film) surrounding the droplets.

159 The evolution of the droplet diameter and temperature are derived from the
160 quasi-steady isolated droplet models, while the evolution of droplet velocity and
161 position are derived from the drag model. By denoting \mathbf{v}_k and \mathbf{x}_k the velocity
162 and position vectors of the k^{th} droplet, the following set of expressions:

$$\frac{d\mathbf{x}_k}{dt} = \mathbf{v}_k \quad (1)$$

$$\frac{d\mathbf{v}_k}{dt} = \frac{1}{\mathcal{T}_{v,k}} (\mathbf{u}(\mathbf{x}_k, t) - \mathbf{v}_k) \quad (2)$$

$$\frac{da_k^2}{dt} = -\frac{a_k^2}{\mathcal{T}_{a,k}} \quad (3)$$

$$\frac{dT_k}{dt} = \frac{1}{\mathcal{T}_{T,k}} \left(T(\mathbf{x}_k, t) - T_k - \frac{B_{T,k} L_v}{C_p} \right) \quad (4)$$

163 are used to track droplet evolution in the computational domain. The scalar
164 $T(\mathbf{x}_k, t)$ and the vector $\mathbf{u}(\mathbf{x}_k, t)$ represent the gas temperature and the gas
165 velocity at the droplet position \mathbf{x}_k . a_k is the diameter of k^{th} droplet, L_v the
166 latent heat of evaporation and C_p the heat capacity of the surrounding gas
167 mixture. The quantities $\mathcal{T}_{v,k}$, $\mathcal{T}_{a,k}$, and $\mathcal{T}_{T,k}$ represent kinetic, evaporation, and

168 heat relaxation time scales:

$$\mathcal{T}_{v,k} = \frac{\rho_l a_k^2}{18C_{D,k}\mu} \quad (5)$$

$$\mathcal{T}_{a,k} = \frac{S_c}{4Sh_c} \frac{\rho_l}{\mu} \frac{a_k^2}{\ln(1+B_{m,k})} \quad (6)$$

$$\mathcal{T}_{T,k} = \frac{P_r}{6Nu_c} \frac{C_l}{C_p} \frac{\rho_l a_k^2}{\mu} \frac{B_{T,k}}{\ln(1+B_{T,k})} \quad (7)$$

169 where ρ_l , C_l are the density and heat capacity of the liquid. μ is the gas viscosity,
 170 S_c and P_r are the Schmidt and Prandtl numbers and Sh_c and Nu_c denote the
 171 convective Sherwood and Nusselt numbers, respectively. The quantities $B_{m,k}$
 172 and $B_{T,k}$ represent the mass and heat transfer numbers (Spalding numbers)
 173 of k^{th} droplet. $B_{m,k} = (Y_v^s - Y_v)/(1 - Y_v^s)$ corresponds to the normalized
 174 mass flux of fuel vapor from the droplet surface Y_v^s to the surrounding gas Y_v .
 175 Its temperature counterpart is $B_{T,k} = (B_{m,k} + 1)^{P_r/S_c \cdot Sh_c/Nu_c} - 1$. For more
 176 information, see [25].

177 The resolution of the reactive multi-species carrier gas phase is based on the
 178 standard set of conservation equations within the low Mach number approxi-
 179 mation. Equations are considered for the mass (i.e., density of the mixture ρ),
 180 momentum components (ρu_i), mass fraction of each chemical species (Y_α), and
 181 sensible enthalpy.

$$\frac{\partial \rho}{\partial t} + \frac{\partial \rho u_i}{\partial x_i} = \dot{m} \quad (8)$$

$$\frac{\partial \rho u_i}{\partial t} + \frac{\partial \rho u_i u_j}{\partial x_j} = -\frac{\partial p_1}{\partial x_i} + \frac{\partial \tau_{ij}}{\partial x_j} + \dot{v}_i \quad (9)$$

$$\frac{\partial \rho Y_\alpha}{\partial t} + \frac{\partial \rho u_i Y_\alpha}{\partial x_i} = \frac{\partial \rho Y_\alpha V_{\alpha,i}}{\partial x_i} + \dot{\omega}_\alpha + \dot{m} \delta_{\alpha v} \quad (10)$$

$$\begin{aligned} \frac{\partial \rho h_s}{\partial t} + \frac{\partial \rho u_i h_s}{\partial x_i} &= \frac{\partial}{\partial x_i} \left[\lambda \frac{\partial T}{\partial x_i} - \rho \sum_{\alpha=1}^N h_{s,\alpha} Y_\alpha V_{\alpha,i} \right] \\ &+ \frac{dP}{dt} + \tau_{ij} \frac{\partial u_i}{\partial x_j} + \dot{\omega}_T + \dot{e} \end{aligned} \quad (11)$$

183 the above system is complemented with the perfect gas equation of state

$$\rho = \frac{PW}{RT} \quad (12)$$

184 ρ , u_i , and T are the density, velocity component in the x_i -direction, and tem-
 185 perature of the mixture, respectively. P and p_1 are the thermodynamic and
 186 dynamic components of pressure. The quantity τ_{ij} is the viscous stress tensor,
 187 $V_{\alpha,i}$ is the i -component of the diffusion velocity of species α , $h_s = \sum_{\alpha=1}^{N_s} Y_\alpha h_{s,\alpha}$
 188 represents the specific sensible enthalpy of the mixture and $h_{s,\alpha}$ is the sensible
 189 enthalpy of species α . The mass fraction of chemical species α is Y_α , $\delta_{\alpha v}$ denotes
 190 the Kronecker tensor, the value of which is unity if the species α matches the va-
 191 por index v , and zero otherwise. The quantities W_α and $W = (\sum_{\alpha=1}^{N_s} Y_\alpha / W_\alpha)^{-1}$
 192 are the molecular weight of species α and the molecular weight of the reactive
 193 mixture, respectively. Finally, R is the universal gas constant.

194 Various source terms are present in the previous set of equations: $\dot{\omega}_\alpha$ and $\dot{\omega}_T$
 195 are related to the chemical reaction processes whereas \dot{m} , $\dot{\mathbf{v}}$, and \dot{e} result from
 196 the two-way coupling between the carrier phase and the spray. $\dot{\omega}_\alpha$ and $\dot{\omega}_T$ cor-
 197 respond to production/destruction rate of chemical species α and the resulting
 198 heat release rate, respectively. They are computed with the chemkin VODE
 199 solver using the n-heptane kinetics of Patel et al. [26], which contains 29 species
 200 and 52 elementary reactions. The terms \dot{m} , $\dot{\mathbf{v}}$, \dot{e} are modifying the mass, mo-
 201 mentum and temperature in the gaseous phase owing to a distribution of the

202 Lagrangian quantities on the Eulerian grid.

$$\dot{m}^{(n)} = \rho_d \frac{\pi}{4} \frac{1}{\mathcal{V}} \sum_k \alpha_k^{(n)} \frac{a_k^3}{\mathcal{T}_{a,k}} \quad (13)$$

$$\dot{\mathbf{v}}^{(n)} = -\rho_l \frac{\pi}{4} \frac{1}{\mathcal{V}} \sum_k \alpha_k^{(n)} a_k^3 \left(\frac{2}{3} \frac{\mathbf{u}(\mathbf{x}_k, t) - \mathbf{v}_k}{\mathcal{T}_{v,k}} - \frac{\mathbf{v}_k}{\mathcal{T}_{a,k}} \right) \quad (14)$$

$$\dot{e}^{(n)} = -C_l \rho_l \frac{\pi}{4} \frac{1}{\mathcal{V}} \sum_k \alpha_k^{(n)} a_k^3 \left(\frac{2}{3} \frac{T(\mathbf{x}_k, t) - T_k - B_{T,k} L_v / C_p}{\mathcal{T}_{T,k}} - \frac{T_k}{\mathcal{T}_{a,k}} \right) \quad (15)$$

203 where \mathcal{V} is a control volume, \sum_k is the sum performed over all droplets affecting
 204 the node n and $\alpha_k^{(n)}$ is the distribution coefficient of the k^{th} droplet source
 205 term on the node n . Considering all the nodes affected by droplet k , it is
 206 necessary to have $\sum_n \alpha_k^{(n)} = 1$ to conserve mass, momentum and energy during
 207 the Lagrangian/Eulerian coupling. The values of $\alpha_k^{(n)}$ are then the normalized
 208 distances between the droplet k and the surrounding nodes.

209 2.2. Configuration

210 As underlined in the introduction, all the simulations are carried out with a
 211 two-dimensional square domain although the evaporation process is kept three-
 212 dimensional. Similarly to [16], this would correspond to a thin slice of a 3D field
 213 with regularly spaced array of droplets in the third direction. A sketch of the
 214 computational domain is shown in figure 2. The side of the domain is 10 mm
 215 long. Both directions are periodic. They are discretized with an isotropic grid
 216 (256×256) keeping grid size $\Delta x = 39.2 \mu\text{m}$. The objective was to reduce as
 217 much as possible the computational cost since this configuration is considered
 218 as an extension of a chemical reactor that will need numerous executions to
 219 underline the impact of various spray physical parameters on gas autoignition.

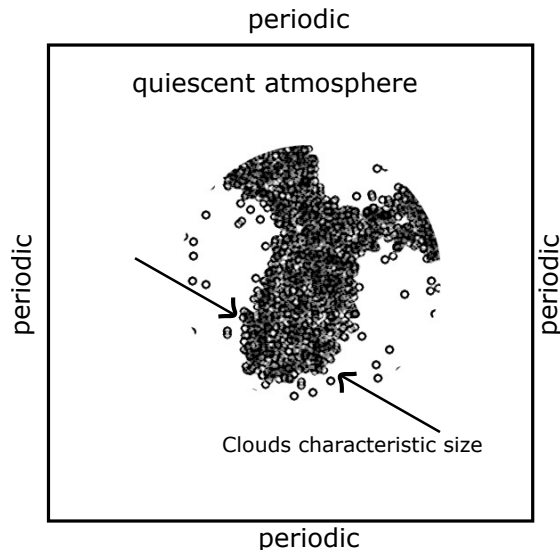


Figure 2: Sketch of the computational domain.

220 The generation of clouds of droplets with a prescribed characteristic size and
 221 density is based on a spectral procedure described by Reveillon [27]. It allows
 222 for generating turbulent velocity and scalar fields along with sprays, the vari-
 223 ation of density of which is prescribed. Some details concerning the control of
 224 the preferential segregation are given here since it is the basis of the extended
 225 reactor configuration. First, it is necessary to define two probability density
 226 functions (PDF): the droplet diameter distribution into the spray $P(a)$ and the
 227 PDF $P(\xi)$ of the spray local density $\xi(\mathbf{x})$, which is the number of droplets per
 228 unit of volume. $P(a)$ may be characterized by any experimental or analyti-
 229 cal shape (Log-Normal, Rosin-Rammler, \dots , [28]) or by a delta function if a
 230 monodispersed spray is considered, which is the case in this paper. Thanks to
 231 the PDF of the spray density, which is prescribed via a β -function [29], it is
 232 possible to generate very different topology of clouds. The mean spray den-
 233 sity $\bar{\xi}$ and $P(a)$ fix the global equivalence ratio of the configuration while the
 234 density variance $\overline{\xi^2}$, necessary to define the β -function, allows for controlling

235 the segregation level $S_L = \overline{\xi^2}/\overline{\xi}(1 - \overline{\xi})$ of the clouds. If $S_L = 1$, a maximum
 236 segregation level is chosen but if $S_L = 0$ the droplets are uniformly and ran-
 237 domly distributed in the domain. A last parameter, δ_c is necessary to prescribe
 238 the characteristic size of the clouds. Examples of generated clouds are shown
 239 in figure 3. On the left, three figures (fig. 3-(a),(b),(c)) shows the same high
 240 level of segregation but the characteristic size of the clouds of droplets has been
 241 diminished from $\delta_c = 2.5 \text{ mm}$ (Fig. 3-(a)) down to $\delta_c = 0.83 \text{ mm}$ (Fig. 3-(c)).
 242 On the right, in figures 3-(a),(d),(e), the same characteristic cluster size is pre-
 243 scribed $\delta_c = 2.5 \text{ mm}$ but the segregation level varies from a highly segregated
 244 spray ($S_L \approx 1$) at the top (Fig. 3-(a)) down to a moderate segregation level
 245 spray ($S_L \approx 0.25$) at the bottom (Fig. 3-(e)). The whole generation procedure
 246 may be found in [27].

247 **3. Reference Case: homogeneous spray autoignition**

248 To identify the spray preferential segregation influence on autoignition, reac-
 249 tivity of a homogeneous mixture with similar thermodynamic conditions inside
 250 a zero-dimensional (0D) reactor has to be first characterized. Consequently, au-
 251 toignition of a two-phase flow n-heptane/air mixture, necessarily homogeneous
 252 is used as reference. This configuration is very close to the reactor used by the
 253 authors in [16]. Therefore, no further detail is given here.

254 Figure 4 depicts two-phase flow n-heptane/air autoignition delay as a func-
 255 tion of the gas initial temperature. The decrease of the autoignition delay with
 256 the increase of the initial gas temperature is reproduced everywhere except in
 257 the *NTC* domain. Even if only a skeletal chemical mechanism was used, the
 258 *NTC* domain is accurately represented, between 850 K and 1050 K in this study
 259 (Fig. 4). Inside this zone, the system reactivity falls down (autoignition delay
 260 becomes longer) while the initial temperature increases.

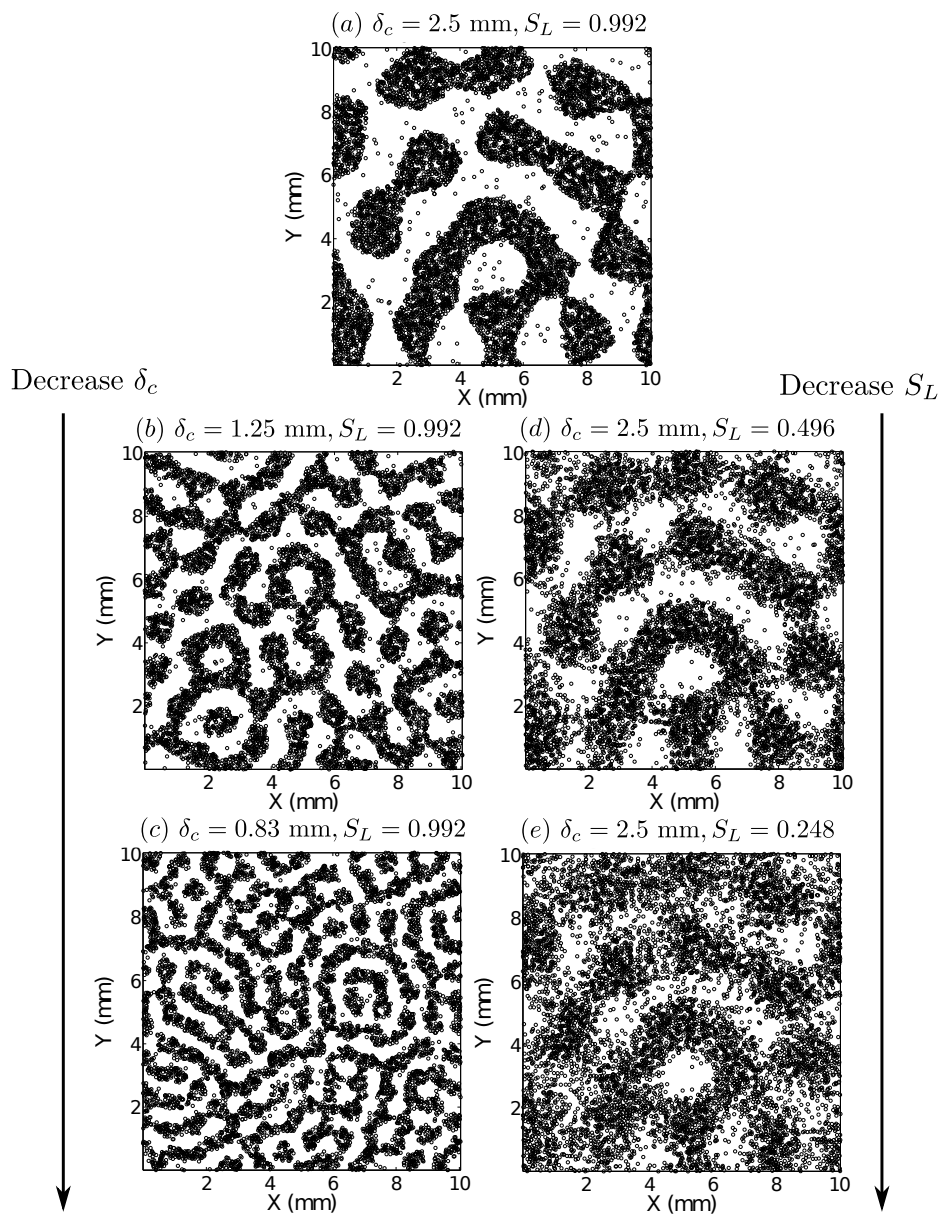


Figure 3: Initial repartition of the clusters of droplets with two prescribed parameters: the characteristic size of the clusters δ_c (left), and the segregation level S_L (right) through the control of the local droplet density.

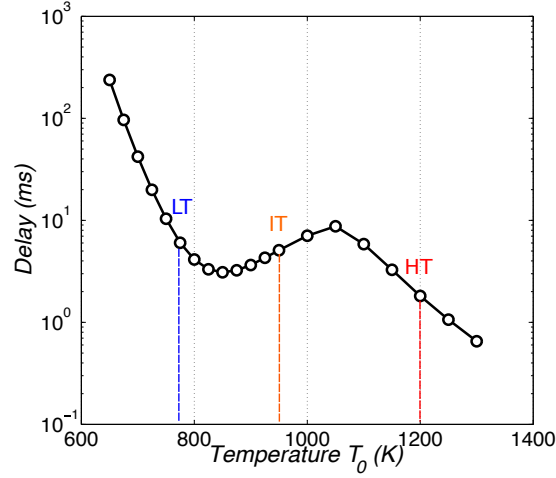


Figure 4: Autoignition delays of a stoichiometric and monodisperse two-phase flow n-heptane/air mixture for different initial temperatures. Initial droplet diameter and pressure are $2 \mu\text{m}$ and 10 bar, respectively.

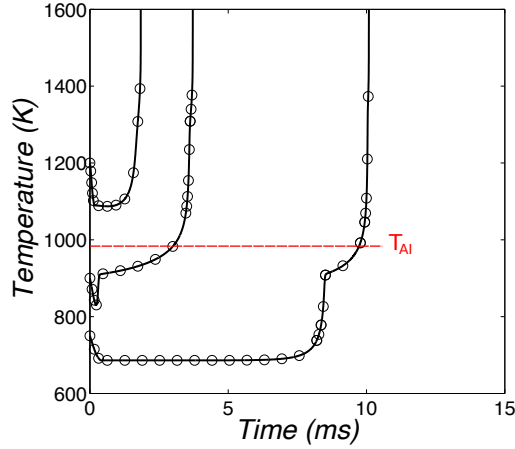


Figure 5: Temporal evolution of temperature for three initial gas temperature ($T_0 = 1200, 900$ and 750 K) in 0D (homogeneous) n-heptane/air two-phase flow reactor. Equivalence ratio is 1, initial droplet diameter and pressure are $10 \mu\text{m}$ and 10 bar, respectively

261 In purely gaseous case, the temperature range of the *NTC* domain is al-
 262 most independent of the fuel/air equivalence ratio but shifts towards higher
 263 temperatures when pressure increases. On the contrary, in former study [16],
 264 authors have shown that in the case of spray autoignition the *NTC* region is

265 simultaneously influenced by the pressure and the equivalence ratio.

266 To analyze the impact of the initial gas temperature on the autoignition
267 process, three distinct initial temperatures have been chosen with respect to
268 the *NTC* temperature range to illustrate the various chemical processes that
269 may be encountered in homogeneous reactors. First, a 'high' temperature level
270 (HT, $T_0 = 1200$ K) has been considered, then an 'intermediate' (IT, $T_0 = 900$ K)
271 and a 'low' (LT, $T_0 = 750$ K) temperature levels.

272 Figure 5 shows the temperature evolution versus time for the three homo-
273 geneous configurations. Whatever the considered case, the liquid vaporization
274 presents a significant temperature decrease. This cooling effect is directly re-
275 lated to the amount of liquid mass receiving the heat flux. In addition of this
276 initial temperature decrease, figure 5 also shows a direct impact of the initial
277 temperature on the autoignition process. In contrast to hot temperature case
278 which is characterized by a one-stage autoignition, the low and intermediate
279 temperature cases present a two-stage autoignition. The first reaction stage
280 increases the temperature of a few hundred degrees. This phase is called '*cool*
281 *flame*'. It is a transition from low or intermediate temperatures to hot tempera-
282 tures that provoke the second stage, which corresponds to the main autoignition.
283 Note that the characteristics of *cool flames* depend both on the initial gas tem-
284 perature and the gas-liquid heat transfers. Figure 5 also shows that the thermal
285 runaway which characterizes the main autoignition, takes place from a fixed
286 temperature threshold whatever the initial temperature.

287 4. Autoignition location

288 4.1. Effect of initial gas temperature

289 To analyze the impact of the initial gas temperature on autoignition in a het-
290 erogeneous system (Fig. 3-(a)), a single spray distribution (monodisperse spray)

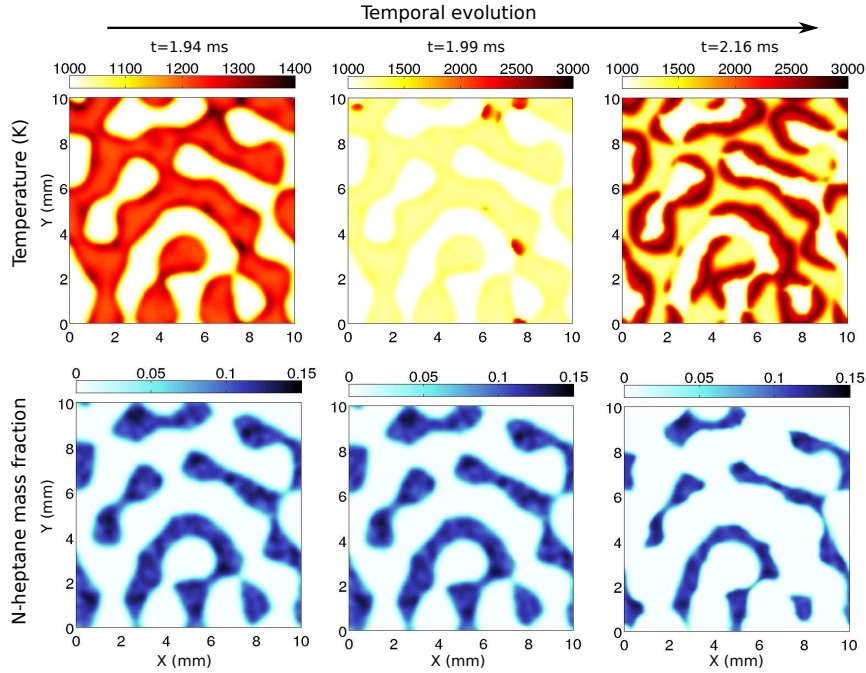


Figure 6: Autoignition of a n-heptane/air mixture with a highly segregated spray (Fig. 3-(a)). Homogeneous initial air temperature: $T_0 = 1200$ K. Top: temperature evolution (K); bottom: n-heptane vapor mass fraction evolution

291 has been used with an initial droplet diameter $a_0 = 10 \mu m$ ($a_0/\Delta x < 0.3$) and
 292 a global equivalence ratio equal to 1. The non-monotonous evolution (*NTC*
 293 region) of the autoignition delay as a function of the temperature and the ap-
 294 parition of *cool flames* for some other specific temperature ranges make anal-
 295 yses more difficult. It is then necessary to vary initial temperature to under-
 296 stand physical phenomena present in the two-phase flows autoignition. This
 297 study is based on the same temperature levels introduced in the reference case
 298 (HT,IT,LT).

299 4.1.1. High temperature (HT)

300 To vaporize, n-heptane droplets capture an important amount of energy
 301 available in the hot gases. Heat exchanges between gas and liquid phases induce

302 localized decreases of the gas temperature that may be seen in Fig. 6-(Top) at
 303 $t = 1.94$ ms, for example. These 'colder' zones are directly related to the pocket
 304 of vapor of fuel that have been formed (Fig. 6-(bottom)). At time $t = 1.99$ ms,
 305 autoignition spots appear in the computational domain (Fig. 6-(middle), see
 306 red spots). They are mostly localized in the preheated areas at the edges of the
 307 pockets of fuel vapor in direct contact with the hot zone.

308 In an initially quiescent environment, the existence of high temperature gra-
 309 dients due to droplet vaporization induces a weak advection process that is
 310 directly linked to the density difference between hot and cold zones. Along
 311 with the molecular diffusion, advection promotes secondary autoignition areas
 312 (Fig. 6-(top), time $t = 2.16$ ms) and leads to a succession of ignition processes.
 313 Fig. 6 shows how, in this specific temperature case, the reactive process starts
 314 from the external frontiers of the vapor pockets and propagates inside. This
 315 process is scrutinized in figure 7-(a) where the gas temperature (top) and the
 316 vapor mass fraction (bottom) evolutions have been plotted. Profiles are based
 317 on the most reactive pocket (Fig. 6, $X = 7.5$ mm and 4.5 mm $< Y < 10$ mm).

318 At time $t = 0.77$ ms, prior to any significant heat release, two areas that
 319 are fuel rich (stoichiometry $Y_v^{st} = 0.0625$) are clearly visible in figure 7-(a)-
 320 bottom with a vapor mass fraction approximately equals to 0.13. These peaks
 321 are directly linked to a 'low' temperature level ($T \approx 970$ K), compared to the
 322 initial gas temperature ($T_0 = 1200$ K), Fig 7-(a)-bottom. As previously stated,
 323 it is the direct consequence of the heat transfer during the droplet vaporization
 324 process. This high temperature gap is far from being negligible, especially from
 325 the autoignition point of view, which is highly non-linear. Note that even if
 326 the local mixture is rich, stoichiometry in the whole reactor is respected from
 327 an average point of view. This kind of non-homogeneous configuration can be
 328 encountered in practical cases since preferential segregation due to the turbulent

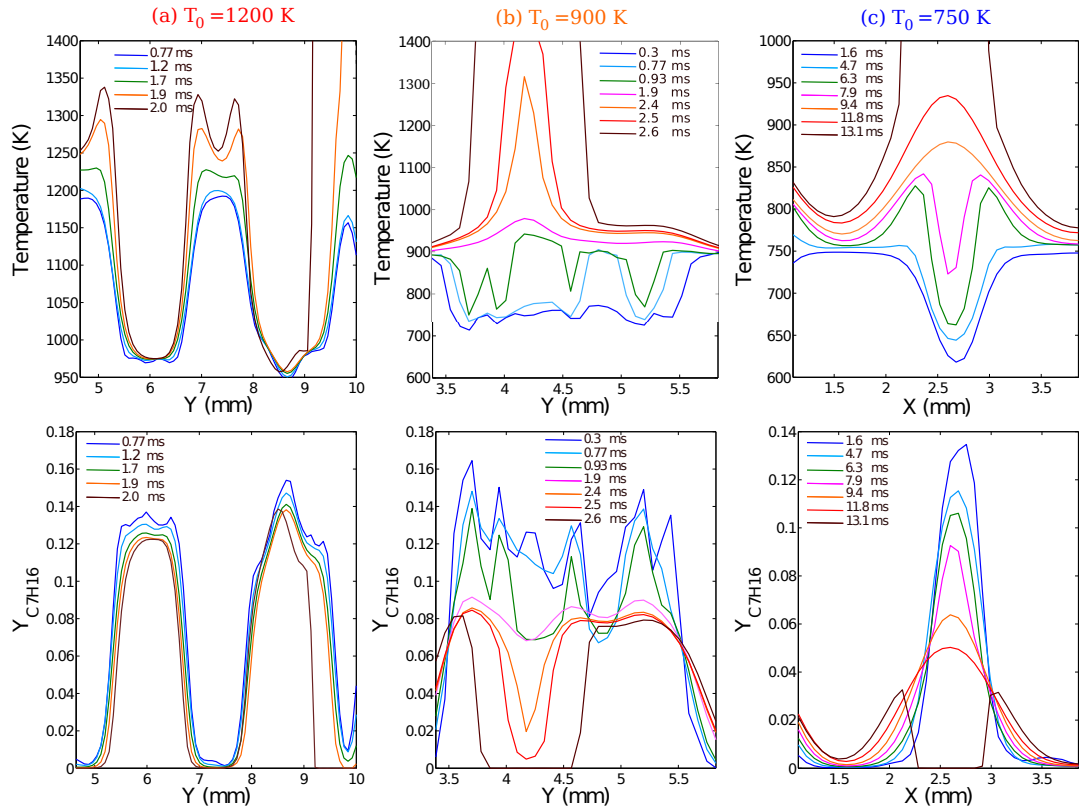


Figure 7: Temporal evolution of the gas temperature (top) and the fuel-vapor mass fraction (bottom) profiles at the most reactive level for three different initial gas temperatures $T_0 = 1200, 900$ and 750 K (left to right).

329 flow may lead to a strongly non-homogeneous droplet dispersion [30].

330 Starting at time $t = 1.2$ ms, a temperature increase may be observed at
331 the periphery of the vapor pockets, mostly on the hot gas regions in the lean
332 side of the vapor mass fraction, see the double temperature peak forming at
333 positions $Y = 6.8$ mm and $Y = 7.8$ mm in (Fig. 7-(a)). Significant heat release
334 represented by peaks of temperature is clearly visible at time $t = 2$ ms and at
335 position $Y = 6.8$ and 7.7 mm, among others. This indicates that the autoigni-
336 tion is controlled by the mixing (diffusion) zones in this quiescent configuration.

337 4.1.2. Intermediate temperature (IT)

338 In the intermediary temperature case ($T_0 = 900$ K), the autoignition pro-
339 cess presents completely different evolution from the previous high temperature
340 ($T_0 = 1200$ K) configuration. This difference is linked to the chemical mecha-
341 nism that characterizes this temperature range. Figure 8-(top) shows snapshots
342 of the temperature obtained at three instants of the autoignition process. The
343 corresponding vapor fields are shown in figure 8-(bottom).

344 Time $t = 0.3$ ms shows first the cooling effect of the vaporization on both
345 the temperature and the vapor fields prior to any significant chemical reaction.
346 Then, at time $t = 1.88$ ms, we observe a general increase of the temperature in
347 all the areas initially occupied by droplets. The temperature increase remains
348 moderated, compared to a full ignition, and it leads to a partial consumption
349 of the fuel vapor. The corresponding vapor and the temperature profiles have
350 been plotted in figure 7-(b). Profiles are extracted from a cut through a single
351 pocket of fuel that corresponds to the most reactive area of the flow (Fig. 8-right
352 ($t = 2.66$ ms), $X = 1.6$ mm and 3.4 mm $< Y < 5.8$ mm).

353 As shown in figure 7-(b), after evaporation, a moderate temperature increase
354 of 150 K is observed inside the fuel rich pockets of vapor at time $t = 0.77$ ms,
355 quickly followed by an elevation of temperature in the domain starting at time

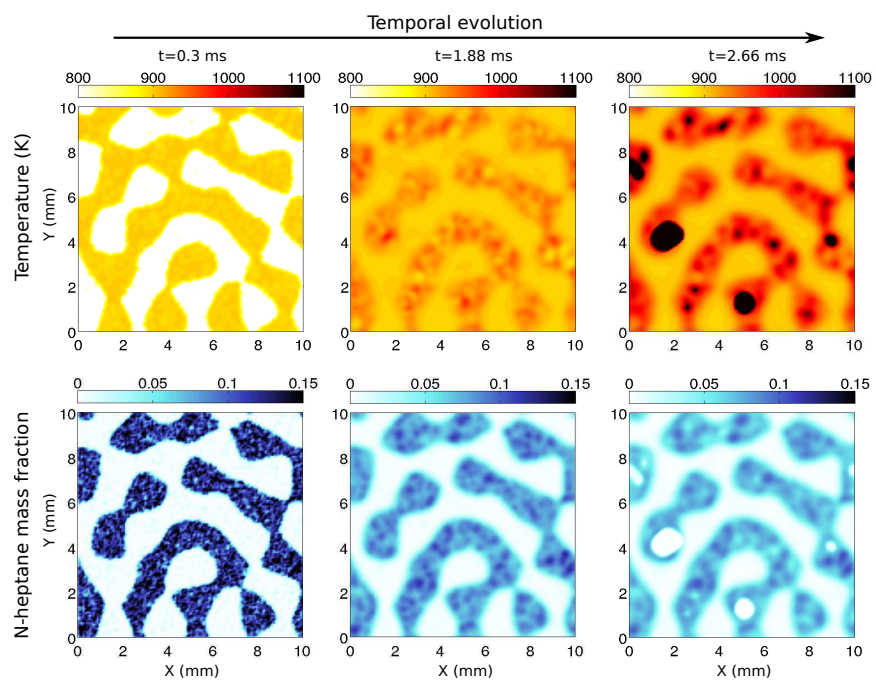


Figure 8: Autoignition of a n-heptane/air mixture with a highly segregated spray (Fig. 3-(a)). Homogeneous initial air temperature: $T_0 = 900$ K. Top: temperature evolution (K); bottom: n-heptane vapor mass fraction evolution

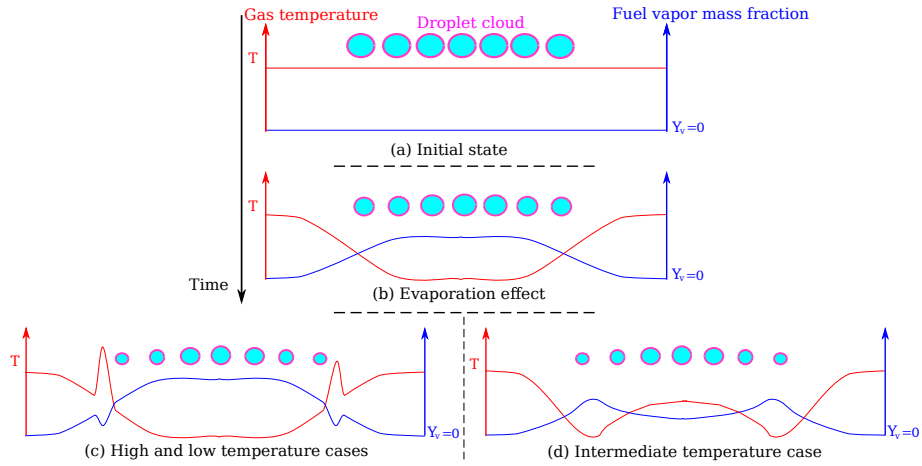


Figure 9: Localization of the most reactive zone: sketch of the evolution of gas temperature and vapor mass fraction profiles through area initially occupied by droplets.

356 $t = 0.93$ ms. It leads, at time $t = 1.9$ ms to an almost uniform temperature
 357 inside and outside the pocket of fuel vapor. In fact, a *cool flame* has appeared
 358 in the pocket of fuel and the heat release due to the *cool flame* has compensated
 359 the heat loss due to vaporization.

360 This first autoignition phase is then followed at time $t = 1.9$ ms by the
 361 complete autoignition of the mixture of the considered area (Fig. 8-(bottom),
 362 $t = 2.66$ ms). The autoignition process occurs in the core of the droplet cluster
 363 in a slightly rich area and then it propagates toward the edges. The fact that the
 364 initial temperature is within the *NTC* range (where the reactivity is inverted
 365 with respect to the temperature increase), autoignition is then promoted not
 366 only by the temperature decreases due to the vaporization, but also by the
 367 amount of fuel vapor available to autoignite (rich mixture). On the other hand,
 368 the main ignition process lead to a total consumption of the vapor of fuel and
 369 'holes' may be observed in figure 8-(bottom), time $t = 2.66$ ms, for example.

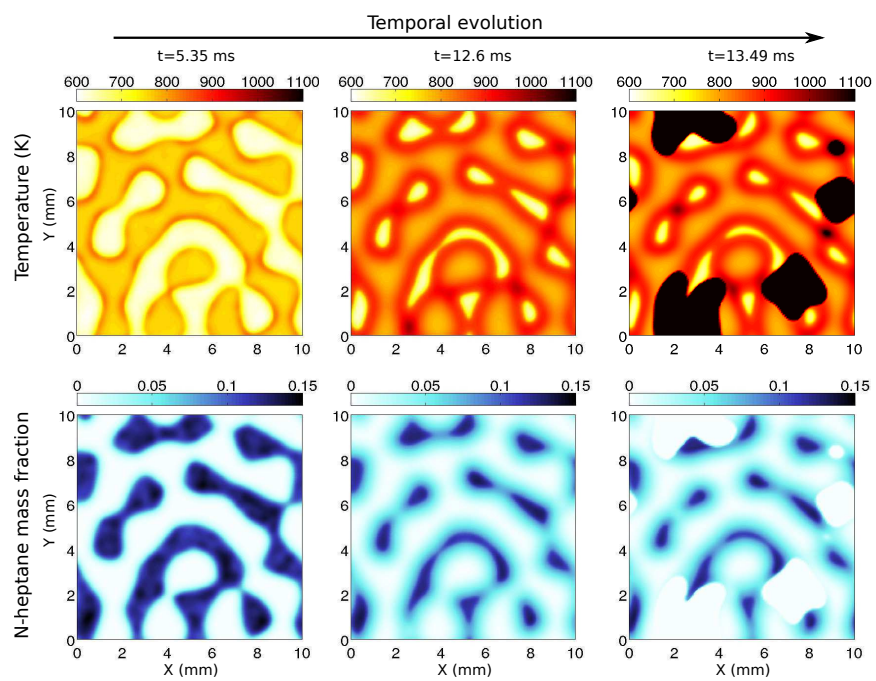


Figure 10: Autoignition of a n-heptane/air mixture with a highly segregated spray (Fig. 3-(a)). Homogeneous initial air temperature: $T_0 = 750$ K. Top: temperature evolution (K); bottom: n-heptane vapor mass fraction evolution

370 4.1.3. Low temperature (LT)

371 In the 'low' temperature case ($T_0 = 750$ K), autoignition of the highly seg-
372 regated spray follows a third scenario compared to the former cases. Figure 10
373 shows snapshots of temperature and vapor fields at three times. Very quickly at
374 $t = 5.35$ ms, it is possible to observe the apparition of *cool flames* at the edges
375 of the areas initially occupied by the droplets (Fig. 10-top-left). Consequently
376 the temperature reaches 850 K around all the fuel vapor pockets. Then this
377 phenomenon propagates toward the center of the pockets by burning partially
378 the available vapor of fuel (Fig. 10-center). Then, the main autoignition occurs
379 at the bridges between close pockets. These bridges are defined by the shorter
380 distance separating two pockets of fuel. In this area, the *cool flames* standing at
381 the boundaries fill more rapidly the pocket inter-distance. Therefore, the heat
382 losses toward cold areas undergone by the *cool flames* is strongly limited since
383 there is less contact with cold zones. Then, the high energy of these 'bridges
384 *cool flames*' promotes the main autoignition. These observations are confirmed
385 by the evolution of the temperature and the vapor profiles. The most reactive
386 area has been selected and analyzed in figure 7-(c) at the position $Y = 0.5$ mm
387 and $1 \text{ mm} < X < 4 \text{ mm}$. Between times $t = 1.6$ ms and $t = 7.9$ ms (Fig. 7-
388 (c)), the *cool flames* apparition at the edge of the cluster are clearly visible
389 ($X = 2.25$ mm and $X = 3$ mm) through a double temperature peak. Later,
390 at time $t = 9.4$ ms the 'bridge *cool flame*', resulting from the union of two
391 initially separated flame, is formed. Then, the energy being high enough, the
392 main autoignition occurs.

393 Figure 9 displays the evolution of the gas temperature and the mass frac-
394 tion profiles from the initial state to the beginning of autoignition. Figure 9-(a)
395 represents the initial state of monodisperse spray with an uniform initial gas
396 composition and temperature, and the figure 9-(b) represents the effect of the

397 vaporization on the temperature and the mass fraction profiles whatever the
398 initial temperature of the gas. We can notice that the strong preferential seg-
399 regation of the droplets generates stratified zones in which the temperature
400 decreases with the amount of fuel vapor. In our case (quiescent flow), these
401 areas are mainly controlled by thermal and molecular diffusion. Since, there are
402 two scenarios concerning the localization of the most reactive zones, it depends
403 on the initial temperature of the gas mixture. The first scenario shown in fig-
404 ure 9-(c) is representative of autoignition of mixtures initially at low and high
405 temperature, in such cases *cool flames* and *main flames* occur at the edges of
406 droplet clusters and propagate towards the center. It should be noticed that,
407 unlike hot temperature cases, our simulation shown that the main autoignition
408 in low temperature case occurs within droplet cloud. The second scenario shown
409 in figure 9-(d) provides an overview of the autoignition of the mixtures initially
410 at intermediate temperature (*NTC* region). In this case the *cool flame* and
411 the main autoignition are favored by the local cooling associated with a high
412 equivalence ratio, therefore, both steps take place inside the cluster.

413 Furthermore, we have observed that the presence of *cool flames* gives rise to
414 autoignition within cloud of droplets. This can be explained by the existence of
415 the autoignition triggering temperature previously discussed in reference homo-
416 geneous case (Fig. 5). Indeed, *cool flames* increase the local temperature of the
417 mixture to the temperature T_{AI} , so that the temperature level become almost
418 uniform in the cluster. This process promotes the autoignition inside the center
419 far from the edges where the thermal and molecular diffusions brake the thermal
420 runaway.

421 4.2. Impact of the size of the clusters

422 As detailed previously, the liquid phase topology in the segregated reactor
423 is characterized by two main parameters: the characteristic size of the clusters

424 δ_c and the droplet segregation level S_L . In practical configurations, these prop-
425 erties are mainly controlled by the atomization process and droplets-turbulence
426 interactions. To begin with, our objective is to study the influence of these
427 parameters on the autoignition but first independently of any turbulent flow
428 (turbulent flow will be considered in the next section). That is why we continue
429 here with non-homogeneous reactor containing a quiescent atmosphere. Three
430 clusters characteristic sizes and three segregation levels have been prescribed.
431 Any configuration may be studied thanks to the method described in [27]. Five
432 configurations (parameters: δ_c and S_L) have been selected, they are visible in
433 figure 3. All other parameters and properties are identical: high temperature
434 environment ($T_0 = 1200$ K, pressure $P_0 = 10$ bar), stoichiometric global equiv-
435 alence ratio and an initially monodispersed spray, the diameter of the droplets
436 being $a_0 = 10 \mu\text{m}$.

437 Let's remember that there is no *cool flame* in 'high' temperature cases. In-
438 deed, the amount of energy is high enough to lead to a quick and complete
439 autoignition without any major influence of the transport phenomena. Thus,
440 the initial temperature $T_0 = 1200$ K has been selected in this analysis in order
441 to observe if there is a direct impact of the spray topology on the main autoigni-
442 tion delay. Figure 11 presents the ignition spots of all configurations. From a
443 purely topological point of view, all ignition spots follow a similar behavior and
444 they appear mostly at the periphery of the clusters whatever their characteristic
445 size.

446 Figure 12-left shows the impact of the variation of the cluster characteristic
447 size on the evolution of the mean temperature (average over the domain) and its
448 deviation. First, the decrease of the cluster size increases the system reactivity
449 leading to an early autoignition. Second, the temperature fluctuations decreases
450 by diminishing the cluster characteristic size. Both observations are the direct

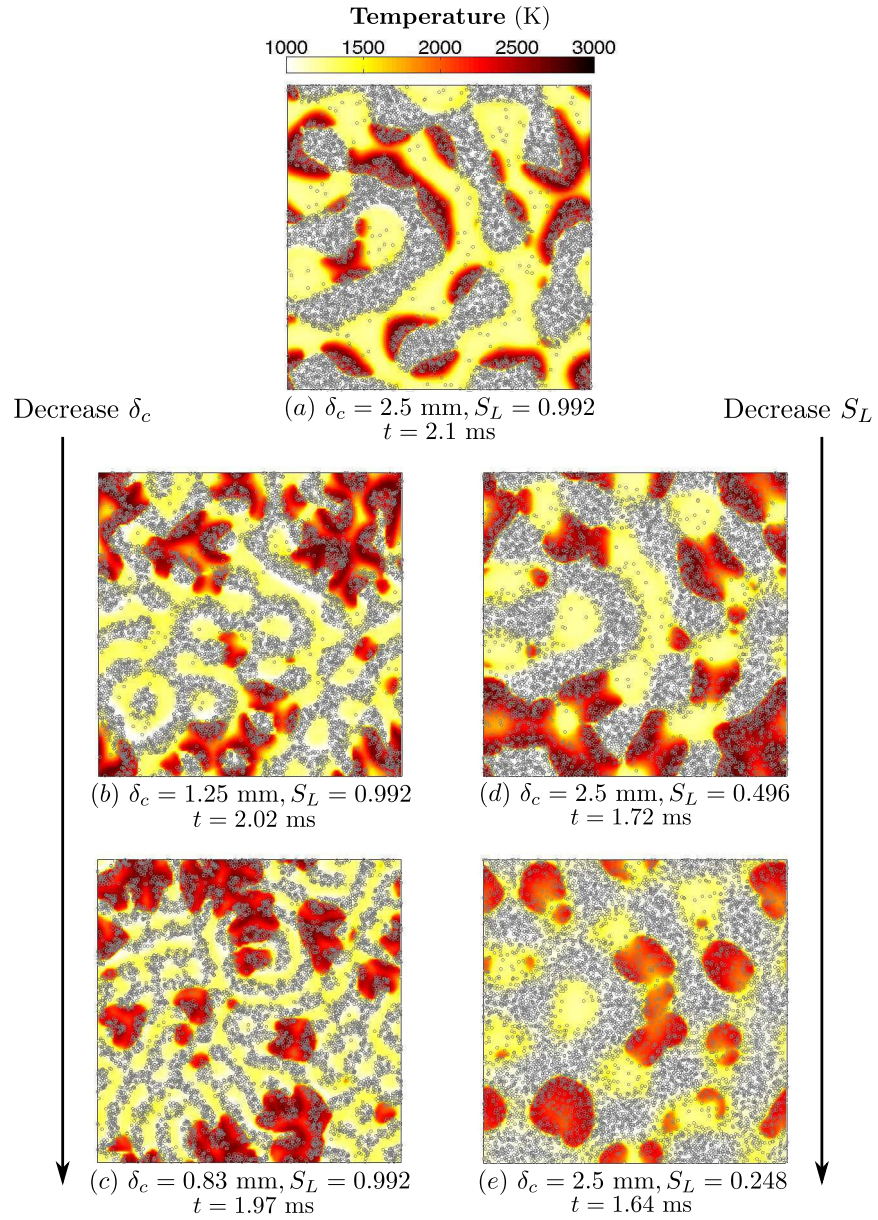


Figure 11: Autoignition of segregated sprays. 'High' temperature case ($T_0 = 1200 \text{ K}$). Left: variation of the characteristic size of the clusters; right, variation of the segregation level.

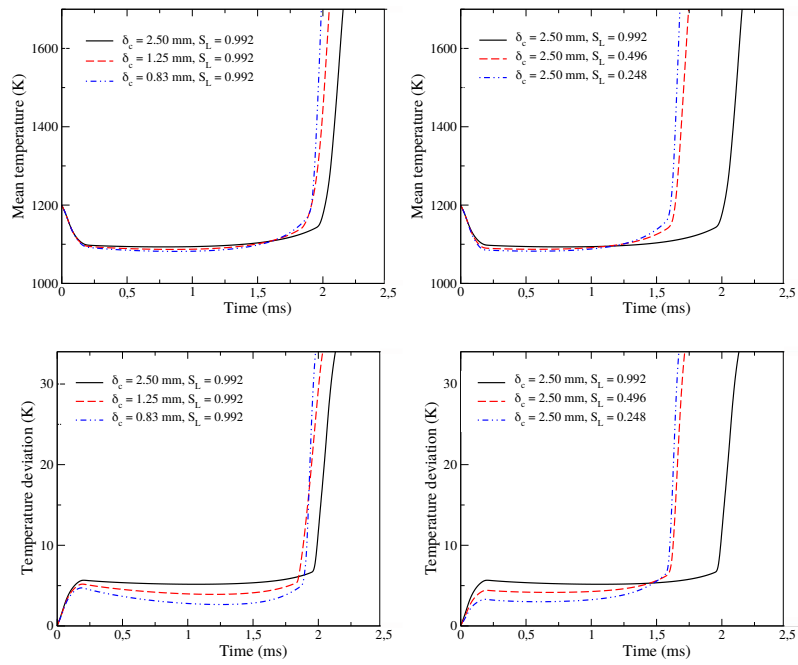


Figure 12: Influence of clusters characteristic size variation (left) and segregation level variation (right) on the mean temperature and the temperature deviation of cases in figure 11.

451 consequence of the increase of the exchange surface between clusters and the
452 hot air, which increases the number of points with the most reactive region.
453 The increase exchange surface also produces more thermal and mass diffusion,
454 and, accordingly a reduction of the local temperature and equivalence ratio
455 fluctuations.

456 The decrease in the segregation level (Fig. 12-right) causes the same effects
457 concerning the temperature deviation temperature and the system reactivity.
458 This tendency remains valid at 'high' and 'low' temperatures. Conversely, if the
459 initial temperature of liquid belongs to the 'intermediate' temperature range,
460 the reactivity can be promoted by a strong local equivalence ratio. In this case,
461 the decrease of the segregation level may delay the autoignition process.

462 **5. Autoignition in turbulent flows**

463 The objective of this section is to add the impact of the turbulent gas fluctu-
464 ations on the most reactive areas and to observe whether there are correlations
465 between the most reactive regions, the scalar dissipation levels and the vorticity
466 intensity. Two-dimensional simulations are performed in a square domain of
467 10 mm by 10 mm. Segregated liquid droplets are embedded in a decreasing
468 homogeneous isotropic turbulence. The turbulent gas velocity field is initialized
469 by specifying a Passot-Pouquet isotropic energy spectrum [27]. The initial ve-
470 locity root mean square u_{rms} is 1.2 m/s, the integral length scale is 0.75 mm
471 and the Kolmogorov length scale is 55.4 μm . The initial droplet velocity was
472 set to zero to isolate the effect of the carrier phase based on the comparison with
473 the previous cases. A 256×256 isotropic Cartesian grid is used ($\Delta x = 39.2 \mu\text{m}$)
474 to capture all the scales of the turbulent flow. Like in the quiescent case, the
475 different initial gas temperatures HT, IT, LT were used: 1200, 900, and 750 K,
476 respectively. The initial pressure for all cases is 10 bar and the global equiva-

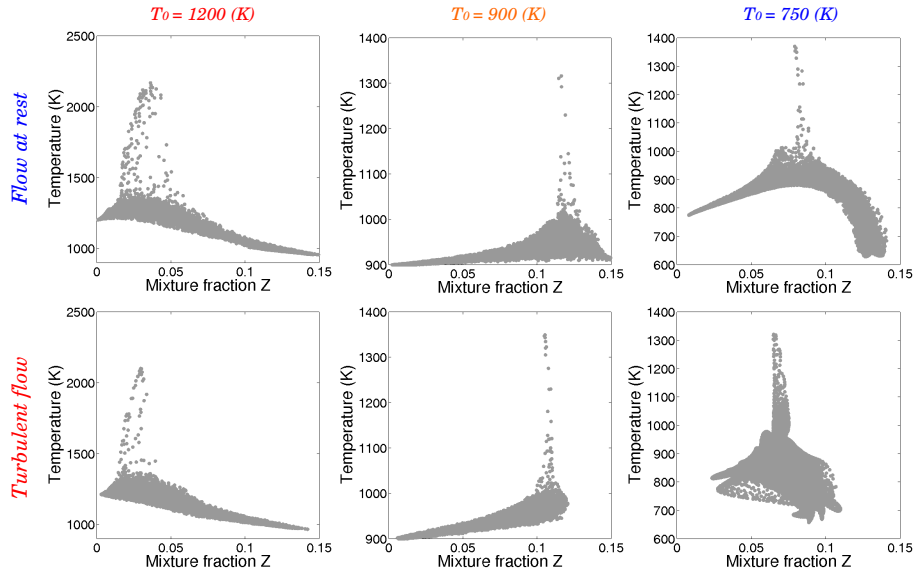


Figure 13: Scatter plots of carrier-gas temperature against mixture fraction for three initial gas temperatures ($T_0 = 1200, 900$ and 750 K) and for two scenarios: (I) flow at the rest (top) and (II) turbulent flow (bottom).

477 lence ratio is 1. The liquid properties are also similar to the previous quiescent
 478 study (initial droplets' diameter and temperature are $10 \mu\text{m}$ and 300 K, re-
 479 spectively).

480 5.1. Most reactive mixture fraction

481 Mixture fraction Z characterizes the mixing between all fundamental ele-
 482 ments of a heterogeneous mixture [31]. Z is equal to 1 in a pure fuel stream
 483 and to 0 in the oxidizer stream. As far as complex chemistry is concerned,
 484 various definitions are possible [32, 33] for the mixture fraction. In this work,
 485 the mixture fraction Z of the flow is based on the vapor of fuel mass fraction.
 486 Indeed, one focuses on the combustion of n-heptane and the definition from [34]
 487 has been selected.

488 In the framework of two-phase flows, it is possible to demonstrate [35] that Z
 489 is not anymore an inert scalar since its time evolution implies the apparition of

490 a mass source term due to droplet vaporization. It is important to characterize
491 Z accurately since it is the key parameter of any combustion model. Therefore,
492 an analysis of the ignition process in the mixture fraction phase space may offer
493 a lot of information for model developments and their applications.

494 One of the key parameter, when studying autoignition, is to determine the
495 value of Z corresponding to the area where the first autoignition spots are
496 detected. It is called the 'most reactive' mixture fraction Z_{MR} .

497 Figure 13 presents scatter plots of the gas temperature against the mixture
498 fraction for two different scenarios: (*I*) flow at rest (Fig. 13- top) and (*II*) tur-
499 bulent flow (Fig. 13- bottom). Both have been applied with the three reference
500 temperature levels (1200, 900, and 750 K). The scatter plots reveal that: (1)
501 ignition occurs first at certain values of mixture fraction (Z_{MR}), the value of
502 which depends on the initial gas temperature. (2) A comparison between the
503 Z_{MR} values with initial turbulence flow and those with flow at rest indicates
504 that the turbulent flow has a weak influence on the most reactive mixture frac-
505 tion at this turbulence level. Note that even if the initial velocity root mean
506 square is the same for all three cases (HT, IT and LT) it reaches different levels
507 at autoignition time since turbulence is decaying and the autoignition delay is
508 different for each case.

509 Indeed, in the 'high' temperature case ($T_0 = 1200$ K) and for both scenarios,
510 figure 13-left shows that the first significant heat release spots are characterized
511 by a most reactive mixture fraction of around 0.025, which corresponds to a
512 gaseous equivalence ratio equals to 0.39. This tendency to ignite in a lean
513 environment is confirmed by the work of Wang and Rutland [8] who observed a
514 similar Z_{MR} , although, quantitative comparisons are not possible since pressure
515 and temperature conditions were different (1 bar, 1700 K, respectively). The
516 gap between the one-phase and two-phase Z_{MR} is directly linked to the presence

517 of the liquid phase that delays autoignition of the rich mixture because of the
 518 proportionally strong decrease of the local temperature. It has been shown by
 519 Bouali et al. [16] and Wang and Rutland [8].

520 For 'intermediate' temperature, ($T_0 = 900$ K, Fig. 13- center), the evolution
 521 of temperature as a function of the mixture fraction is different. The first main
 522 heat release spots are localized in rich areas ($Z_{MR} = 0.115$). This Z_{MR} corre-
 523 sponds to a main ignition. It follows a *cool flame* creation which is localized in
 524 a Z_{MR} area that is less rich (around the stoichiometry).

525

526 As far as 'low' temperature ($T_0 = 750$ K) is concerned, the most reactive
 527 mixture fraction corresponding to the *cool flame* autoignition is very small. It
 528 explains why the *cool flames* are mostly located at the edge of the fuel pockets.
 529 Indeed, it needs only a small amount of fuel to ignite since it is favored by
 530 relatively high local temperature levels. On the contrary, the main autoignition
 531 spots are situated in slightly rich areas (Fig. 13- right) where $Z_{MR} = 0.08$ (to
 532 be compared with the stoichiometric value: $Z^{st} = 0.0625$), *i.e.*, where the fuel
 533 vapor quantity is sufficient to start-up the main autoignition.

534 Figure 14 shows instantaneous snapshots of early autoignition of tempera-
 535 ture, mixture fraction, vorticity and scalar dissipation:

$$\chi = \left(\frac{\partial Z}{\partial x} \right)^2 + \left(\frac{\partial Z}{\partial y} \right)^2, \quad (16)$$

536 at time $t = 1.88$ ms, $t = 2.59$ ms and $t = 7.08$ ms for the reference temper-
 537 atures, HT, IT, LT, respectively; the first autoignition sites embedded in the
 538 temperature fields (black spots) are shown in the other fields by iso-contours
 539 (red color). The first autoignition spots are located in areas of low scalar dis-
 540 sipation rate, where mixture fraction gradients are relatively small and where
 541 there is an adequate mixing level. These zones appear also if the turbulence

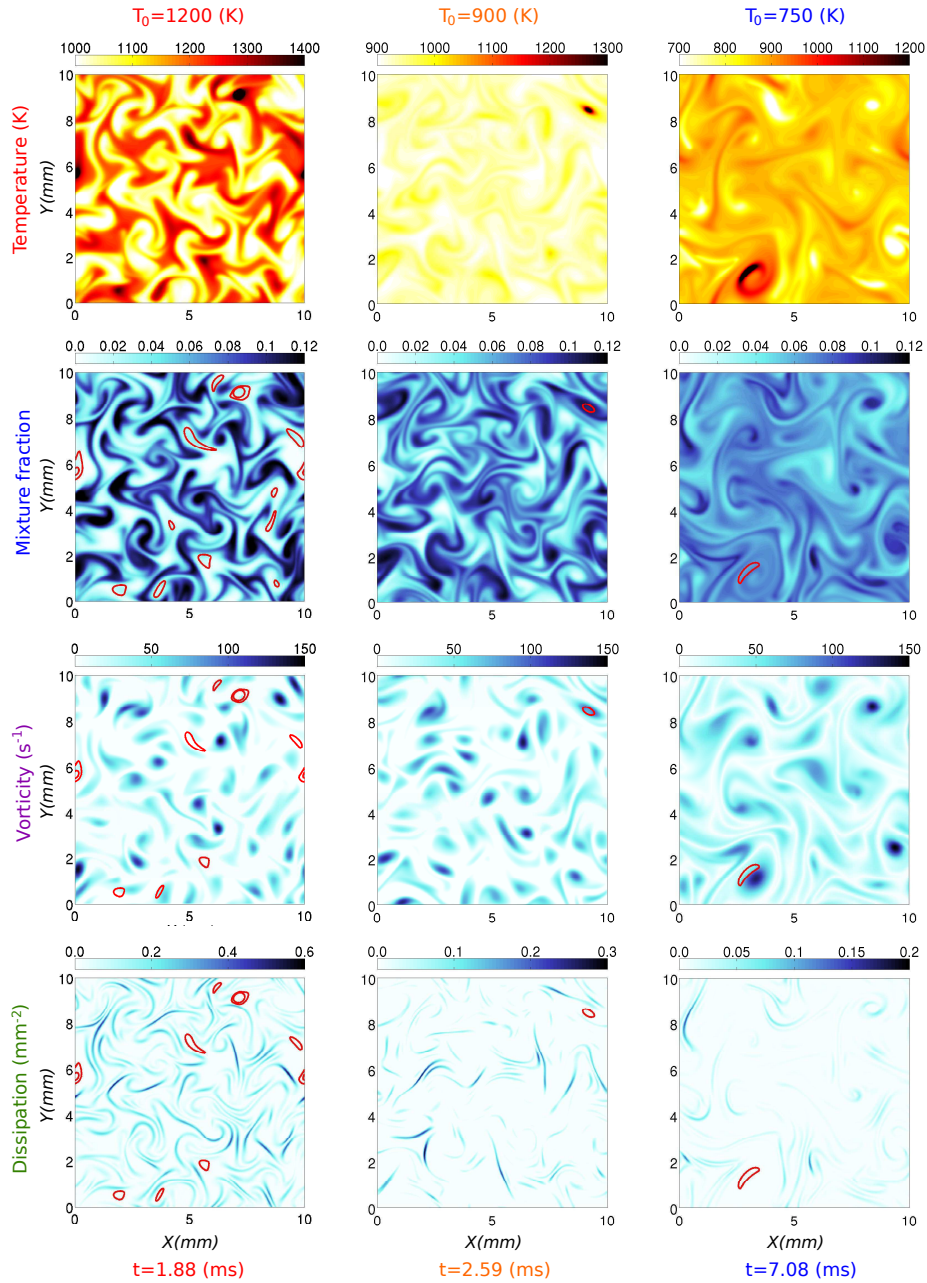


Figure 14: Snapshots of temperature (K), mixture fraction, vorticity (s^{-1}) and scalar dissipation (mm^{-2}) (top to bottom) for three initial gas temperatures $T_0 = 1200, 900$ and 750 K (left to right) at the moment of the ignition. Red iso-contours indicate the most reactive regions.

542 intensity and the mean value of strain rate are high. This result fully matches
 543 the outcomes in [9, 1, 36, 3], where a simpler chemistry models for n-heptane
 544 was used, and in [7, 8] where a skeletal chemical mechanism for same fuel was
 545 considered. Studies [37, 38] using a reduced chemical mechanism for hydrogen
 546 confirm this remark. This observation is valid whatever the initial gas temper-
 547 ature. This finding proves the importance of fluid dynamic phenomena in the
 548 autoignition process.

549 Unlike scalar dissipation rate, the existence of a correlation between vorticity
 550 magnitude and the first autoignition areas is not verified as figure 14 shows that
 551 the first autoignition spot may occur at low (HT case), medium (IT case) and
 552 strong (LT case) vorticity magnitude.

553 *5.2. Average temperature over the Extended Reactor*

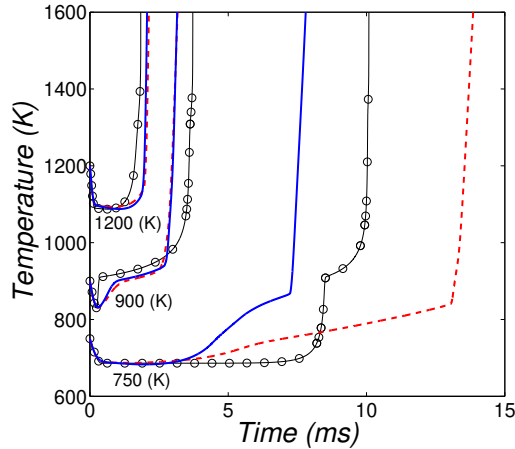


Figure 15: Average over the spatial domain of the gas temperature for spray autoignition configurations. Three initial gas temperature are compared ($T_0 = 750 K$, $900 K$ and $1200 K$) for three initial scenarios: (I) segregated reactor with flow at rest, red dashed lines ; (II) segregated with turbulent flow, blue solid line; (III) homogeneous droplet distribution (0D reactor), circles with black solid line.

554 The influence of the initial gas temperature on the location of the first au-
 555 toignition spots in a segregated two-phase flow has been studied in the previous

556 part. To complete this study, it is interesting to analyze the impact of segre-
557 gated spray on the mean temperature in the reactor. Our objective is to extend
558 classical autoignition studies of homogeneous mixtures or two-phase flows in 0D
559 reactors to non-homogeneous configuration with droplet dispersion. Therefore
560 the averaged output of our multidimensional reactor has to be compared to a
561 0D two-phase flow autoignition reactor. The input of both reactors are the same
562 except for the segregation of the droplets. Note that it has been checked that
563 our multidimensional reactor used with a uniformly distributed spray gives ex-
564 actly the same result than a usual 0D reactor. Details concerning autoignition
565 of homogeneously distributed two-phase flows may be found in [16].

566 The volume-averaged gas temperature of three scenarios is shown in fig-
567 ure 15. The initial temperatures ($T_0 = 750$ K, 900 K and 1200 K) and stoi-
568 chiometric global mean equivalence ratio of the three scenarios that have been
569 compared are similar. The only difference lays in the high segregation level of
570 the segregated reactors and the nature of the flow: scenarios (*I*) and (*II*) cor-
571 respond to a segregated reactor, (*I*) with flow at rest and (*II*) with turbulent
572 flow; scenario (*III*) is the homogeneous reactor (0D).

573 The 'low' temperature case with segregated scenarios (*I*) and (*II*) generates
574 a first exothermic chain of reactions that occurs earlier ($t = 5$ ms) than in
575 scenario (*III*) ($t = 8$ ms), the homogeneous reactor. It corresponds to the
576 apparition of the *cool flame* at the edges of the pocket of fuel. In segregated
577 droplets, ignition occurs at an earlier stage since (1) in areas where droplets are
578 present, the mass of vapor increases more rapidly and (2) outside these areas,
579 the temperature remains at its initial level whereas it decrease everywhere in the
580 homogeneous scenario (*III*). Therefore thermochemical conditions to initiate a
581 reaction (a *cool flame*, given the temperature range) appear more quickly in the
582 segregated scenarios.

583 Once the *cool flame* occurs in the homogeneous scenario, the mean temper-
584 ature reaches very quickly 900 K. It induces very quickly the main autoignition
585 process ($t = 10$ ms). On the other hand, temperature elevation in the segre-
586 gated scenarios is generally slower because the temperature field resulting from
587 the vaporization is also segregated and its evolution depends not only on lo-
588 calized reactive processes but also on diffusion and advection phenomena, the
589 characteristic times of which are larger. Note that for scenario (*II*), even if the
590 elevation of global *cool flame* temperature is relatively slow, it is much quicker
591 than scenario (*I*). This is due to the turbulent flow that promotes the autoigni-
592 tion propagation.

593 For 'low' initial carrier-gas temperature, the figure 15 shows that the mixture
594 heterogeneity may decrease or increase the reactivity of the system depending
595 on the scenario. Indeed, the main autoignition is governed by segregation and
596 transport phenomena. If the *cool flame* delay resulted from the homogeneity
597 is greater than the elevation temperature delay caused by the heterogeneity,
598 the segregation increases the system reactivity (scenario (*II*)). Otherwise, the
599 system reactivity decreases (scenario (*I*)).

600 In the 'intermediate' temperature range ($T_0 = 900$ K), the general behavior
601 of autoignition is the same but its characteristics change: (1) we see quick de-
602 crease of the mean temperature to vaporize the liquid phase. The same delay is
603 necessary (less than 1 ms) in the three scenarios. (2) Then a *cool flame* appears
604 in all reactors. It leads to a quick increase of the mean temperature in the
605 homogeneous case while it is more gradual in the segregated ones. (3) Finally,
606 autoignition occurs with delays that are slightly different. Indeed, ignition of
607 segregated scenarios occurs half a millisecond earlier (2.5 ms versus 3 ms). It
608 is directly linked to the topology of the flow, because, in this case, ignition oc-
609 curs in the core of the pockets of fuel whereas in the two other cases (low an

610 high temperature), it is localized at the edges and much more dependent on
611 temperature/vapor diffusion.

612 At high temperatures ($T_0 = 1200$ K), the heterogeneity of the mixture
613 delays slightly the autoignition, despite the fact that the segregation creates
614 favorable autoignition sites. These areas have suffered of reduced heat losses
615 and their mixture fractions are equal to the most reactive mixture fraction for
616 $T_0 = 1200$ K ($Z_{MR} = 0.025$). However, these sites are regions where strong
617 heat and mass exchanges exist, thus they are characterized by high scalar dissi-
618 pation rates that delays ignition. In practice, the first autoignition site located
619 in the most reactive region in the segregated case appears at time $t = 1.88$ ms,
620 while the autoignition in a homogeneous reactor (no scalar dissipation region)
621 of a stoichiometric mixture occurs at time $t = 1.84$ ms.

622
623 As a first conclusion, it is first possible to state that the presence of a seg-
624 regated spray delays autoignition, but only with some specific conditions, an
625 earlier start is possible. Second, the spray segregation does not suppress the
626 *cool flame* process. On the contrary, various topologies of *cool flames* have been
627 observed and they strongly impact the flow evolution.

628 **6. Conclusion**

629 This study extend the chemical reactor configuration to investigate the effect
630 of preferential segregation on autoignition of n-heptane/air two-phase flow. An
631 Eulerian description is employed to solve the carrier-gas phase and a Lagrangian
632 approach is used to track the liquid-fuel droplet properties. Two-way coupling
633 interaction is considered through the exchange of mass, momentum, and energy
634 between the carrier-gas phase and the dispersed phase. To describe the chemical
635 reaction paths, a chemistry mechanism with 29 species and 52 reactions was

636 chosen. The effects of initial gas temperature (750, 900 and 1200 K) and
637 droplet segregation were examined. The main findings are:

- 638 1. The presence of a segregated spray mainly delays of the autoignition, but
639 only under some specific conditions, an earlier start is possible.
- 640 2. The spray segregation does not suppress the *cool flame* process. On the
641 contrary, various topologies of *cool flames* have been observed and they
642 strongly impact the flow evolution.
- 643 3. Important impacts of initial carrier-gas temperatures on the localization
644 of the first autoignition sites has been identified. This is the consequence
645 of non-monotonic dependence of Z_{MR} on initial gas temperature.
- 646 4. In agreement with previous literature, this study shows that first autoigni-
647 tion spots always occur at locations with lower scalar dissipation. The
648 strong scalar dissipation can even control the ignition more than Z_{MR}
649 parameter.
- 650 5. For the level of turbulence intensity chosen for this study, the values of
651 Z_{MR} are weakly influenced by the turbulent flow and no correlation has
652 been established between the first autoignition site and the vorticity mag-
653 nitude. Indeed, the autoignition may occur at low as well as high vorticity
654 magnitude.

655 **7. Acknowledgments**

656 The authors acknowledge the financial support from IFPEN. A part of this
657 work has been performed using high-performance computing (HPC) resources
658 from GENCI (Grand Equipement National de Calcul Intensif)-[CINES/IDRIS]
659 under Grant A0052B07456.

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