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# Influence of interface conditions on laser diode ignition of pyrotechnic mixtures: application to the design of an ignition device

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#### **Abstract**

This paper treats of numerical modelling which simulates the laser ignition of pyrotechnic mixtures. The computation zone is divided into two fields. The first is used to take account of the heat loss with the outside. It can represent an optical fibre or a sapphire protective porthole. The second field represents the reactive tablet which absorbs the laser diode's beam. A specific feature of the model is that it incorporates a thermal contact resistance *Rc* between the two computation fields. Through knowledge of the thermal, optical and kinetic properties, this code makes it possible to compute the ignition conditions. The latter are defined by the energy  $E_{50}$  and the time  $t_i$  of ignition of any pyrotechnic mixture and for various ignition systems.

This work was validated in the case of an ignition system consisting of a laser diode with an optical lens re-focussing system. The reactive tablet contains 62% by mass of iron and 38% by mass of KClO<sub>4</sub>. Its porosity is 25.8%. After an evaluation of the laser's coefficient of absorption, the variations of the ignition parameters  $E_{50}$  and  $t_i$  are studied as a function of the thermal contact resistance  $R_c$ . Temperature profiles are obtained as a function of time and for various values of the thermal contact resistance *Rc*. More fundamental observations are made concerning the position of the hot spot corresponding to priming. From this study, which concerns the heat exchange between the two media, several practical conclusions are given concerning the design of an ignition device. By evaluation of the thermal contact resistance *Rc*, comparison with test results becomes possible and the results of the computations are in reasonable agreement with the test measurements. 2002 Éditions scientifiques et médicales Elsevier SAS. All rights reserved.

*Keywords:* Laser ignition; Contact resistance; Pyrotechnic mixtures; Thermal ignition model

## **1. Introduction**

The ignition, by means of a laser source, of the explosive reactions of energetic materials, is interesting from a fundamental and practical point-of-view.

Laser diodes are of particular interest because of their very high reliability and their exceptional energy outputs. Because of their small size  $(4 \times 4 \times 1 \text{ cm}^3)$ , they could be used as an ignition system in cars for air bags or seatbelt pre-tensioners. One of the best advantages of laser diodes concerns the absence of sensitivity to electromagnetic disturbance. Furthermore, the power deposited is perfectly well-known as a reliability criterion.

The purpose of this paper is to model the thermal ignition phenomenon of energetic materials, in the case of ignition by a laser diode, with power-flux densities which can be up to 10 kW·cm−2. A comparison between numerical modelling and the results of our experiments is then carried out.

Previous modelling was carried out for such an application, but the model was limited by a restricting hypothesis. For example, Gillard et al. [1] presented a two-dimensional model where only one field (the pyrotechnic mixture) was considered. Physical parameters, such as thermal conductivity, density and heat capacity, were assumed to be constants and a zero-order kinetic law was used.

The new model presented here removes the restriction of the preceding model. This model ought to make it possible to rapidly compute the duration and energy of a pyrotechnic mixture's ignition. The computation zone is divided into

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## **Nomenclature**



two fields. The first can represent an optical fibre (in the case of a direct contact ignition system) or a protective porthole (ignition using lenses). The second computation field represents the reactive pyrotechnic mixture which absorbs the laser beam. The absorption depth cannot be measured, so an approximate computation is proposed to evaluate it. By taking into account these two fields, it is possible to consider the energy losses in the optical fibre or protective porthole. The model incorporates a thermal contact resistance between the two fields so as to take account of a thin film of air contained in the rough areas. Because it is very difficult to measure this parameter, an approximate computation method is proposed. By the evaluation of the thermal contact resistance and of the coefficient of absorption, it is possible to validate the computation code by comparing the computed results with those from the tests carried out in our laboratory. The influences of the thermal contact resistance,  $Rc$ , and of the coefficient of absorption,  $\mu$ , have been studied. From this study, the characteristics of the ignition are explained and several practical conclusions are given concerning the design of a laser diode thermal ignition device.



## **2. Test set-up**

The experimental set-up, used in this study, has been described by Radenac [2]. A laser diode, whose beam is channelled by two lenses, is re-focused onto the pyrotechnic tablet to be initiated. A sapphire porthole, juxtaposed against the tablet, protects the optical system during the explosive reaction.

All of the componements presented in Fig. 1 are fixed on a rail that is integral with an optical table, which facilitates the alignment of the various components.

The power output of the diode used is 7 W and its wavelength is 820 nm. The pulse time can be adjusted from 5 µs to 300 ms. The laser beam emerges at the end of a sheathed optical fibre. The fibre is fixed on a holder whose position is adjusted with a micrometer plate located at its base. The initiation time measured with an infrared sensor [2]. A filter was chosen which does not transmit the wavelength emitted by the laser diode; so that the sensors only perceive the radiation emitted by the pyrotechnic composition. This makes it possible to measure the initiation time even when the tablet is irradiated by the laser. The energy deposited on the surface of the tablet is measured by



Fig. 1. Experimental set-up.

means of an R752 type pyroelectric joulemeter supplied by Digirad. The energy is proportional to the deposition time because the power supplied by the diode is strictly constant.

## **3. Modelling**

The test set-up was used for the ignition of several mixtures [3]:

- $Zr/KClO_4$  [2,4];
- $Zr/PbCrO<sub>4</sub>$  [2,5];
- Fe/KClO<sub>4</sub> [6,7].

Because of the mixture's kinetic and optical parameters are known, numerical simulation can be tested with the  $Fe/KClO<sub>4</sub>$  mixture. The mixture consists of 62% iron (by mass) and 38% of  $KClO<sub>4</sub>$ , and the porosity of the tablet thus obtained is 25.8%.

So as to model all of the thermal phenomena during ignition, the sapphire porthole is incorporated in the model. There are then three interfaces, the most significant of which is the one between the porthole and the pyrotechnic composition. It will be demonstrated below, that this interface governs the dynamics of ignition. An initial model (the results of which are not presented here) assumed perfect contact between the pyrotechnic composition and the porthole, but the thresholds computed were then far too great. In the case of the lowest power outputs supplied by the laser, ignition was even rendered impossible. That is explained by a very great dissipation of the heat in the sapphire porthole. In fact, the tablet is placed against the porthole but a thin layer of air remains trapped, forming an insulating medium which reduces the energy loss (see Fig. 2(a)). The consequence of this insulator is that the face of the porthole (tablet side) is not at the same temperature as the surface of the pyrotechnic mixture. The continuity of the temperature is provided by the thin space between the two faces. In this model, the phenomenon is treated, on a macroscopic scale, by a discontinuity of the temperature at the interface. A thermal contact resistance is thus introduced on this interface. The latter is defined by the ratio:

$$
R_c(T) = \frac{e}{\lambda_{\text{air}}(T)}\tag{1}
$$

where *e* is the equivalent air thickness defined by Fig. 2(a) and (b), and  $\lambda$  the thermal conductivity of the gas which fills the interstices in the interface. The resistance *Rc* represents this air thickness. At the porthole/tablet interface, the limit conditions will be determined by the equality of the flows of the heat. In Fig. 3, the modelled fields and the interfaces taken into account for the computations are shown. The finite-difference method meshing is smaller on the porthole/tablet interface than on the pyrotechnic tablet. The field for the sapphire porthole is meshed with a constant space pitch.

#### *3.1. Basic equations*

To deal with the thermal phenomenon of laser ignition, the condensed phase ignition model is used [8]. Gillard has shown that radial diffusion is negligible compared with axial diffusion along the centre line of the laser beam [1]. Thus the ignition model is based on the equation of the heat developed in a single dimension of space.

The model takes account of the heating by the laser (for a laser beam perpendicular to the surface) inside the tablet (considered as reactive). The heat losses in the porthole and the thermal contact resistance are also considered.

The illustration of the equations, fields and interface is given in Fig. 3 and we discuss first the air/porthole interface.

#### *3.2. Field I: The sapphire porthole*

Sapphire being a transparent material for the wavelength (820 nm) of the diodes used, there is no energy absorption in this medium. The only losses are due to the reflections on each of the faces. The formulation of Fourier's equation is then reduced to its simplest expression:

$$
\frac{\partial T}{\partial t} = \frac{1}{\rho_h(T)C p_h(T)} \frac{\partial}{\partial z} \lambda_h(T) \frac{\partial T}{\partial z}
$$
 (2)





## *3.3. The air/porthole interface*

The heat flux continuity equation is given by:

$$
h_e(T_{\rm EX} - T_{z=0}) = -\lambda_h \frac{\partial T}{\partial z}\Big|_{z=0}
$$
\n(3)

## *3.4. Field II: The pyrotechnic tablet*

Assuming  $\alpha = \varepsilon$  (Kirchoff's law), Fourier's equation in our case is written :

$$
\frac{1}{\frac{\partial T}{\partial t}} = \frac{1}{\frac{1}{\rho_c(T)C\rho_c(T)}} \frac{\frac{\partial T}{\partial z}}{\frac{\partial T}{\partial z} \lambda_c(T) \frac{\partial T}{\partial z}}
$$
\n
$$
+ \frac{1}{\frac{Q}{C\rho_c(T)} \left[1 + (n-1)K_0 \int_0^t e^{-\frac{E_a}{RT(t)}} dt\right]^{\frac{n}{1-n}} K_0 e^{-\frac{E_a}{RT}}}
$$
\n
$$
+ \frac{1}{\rho_c(T)C\rho_c(T)} \frac{P}{S} e^{-\mu z}
$$
\n(4)

In relation  $(4)$ , the term II is the thermal diffusion written with a possible variation of the thermal parameters. The term III represents the heat released by the chemical reaction. A kinetic law with an *n*th order of reaction was considered. The last term (IV) is the in depth absorption of the laser beam which is equivalent to a volumetrical heat source.

The specific heat capacity  $C_p$  and the density  $\rho$  are computed according to the porosity *ε* and the proportions by mass of the constituents of the mixture by the relationships (5) and (6)

$$
C p_c = X_{\text{air}} C p_{\text{air}} + (1 - X_{\text{air}}) \sum_{i=1}^{N} X_i C p_i
$$
 (5)

$$
\rho_c = \varepsilon \rho_{\text{air}} + (1 - \varepsilon) \rho_{\text{solid}} \tag{6}
$$

where

$$
\rho_{\text{solid}} = \left[ \sum_{i=1}^{N} \frac{X_i}{\rho_i} \right]^{-1} \tag{7}
$$



 $\overline{1}$   $\overline{2}$   $\overline{2}$   $\overline{2}$ 





The thermal conductivity,  $\lambda_c$ , was computed according to the porosity *ε* and proportions of the constituents of the mixture using the relationship presented by Tye [9].

The kinetic parameters  $E_a$ ,  $K_0$  and  $n$  are listed in Table 1. They are used as input data for the model presented here. By using these data and incorporating the kinetic law, it is possible to determine, as a function of time, the temperature of the reactive medium for each position in space. The advancement  $\eta$  of the pyrotechnic reaction is given by:

$$
\frac{d\eta}{dt} = K_0 e^{-E_a/(RT(1-\eta))^n}
$$
(8)  

$$
\eta(T) = 1 - \left[1 + (n-1)K_0 \int_0^t e^{-E_a/(RT(t))} dt\right]^{1/(1-n)}
$$
(9)

 $\boldsymbol{0}$ 

 $\overline{\phantom{a}}$ 

with

$$
\int_{0}^{t} e^{-E_a/(RT(t))} dt < \frac{-1}{K_0(n-1)}
$$
 if  $n < 1$ 

The absorption depth *z*ab is defined by Beer–Lambert's law ( $P(z) = P(0)e^{-\mu z}$ ), thus  $z_{ab} = 4.605/\mu$  if one considers the ratio  $P(z)/P(O) = 1/100$ .

## *3.5. The window/tablet interface*

The heat transfers for this interface are very significant in understanding the physical energy transfer phenomenon. These exchanges are done by dissipation of heat from the surface of the tablet inwards and from the surface of the tablet towards the porthole, in a more or less efficient manner according to the value of the thermal contact resistance. At this interface, there is a discontinuity of the temperature which necessitates the writing of an equation for the heat fluxes in each side of the interface. The following two relationships are obtained, the resistance *Rc* having been defined in (1):

*Porthole side*:

$$
-\lambda_h \frac{\partial T}{\partial z}\Big|_{z=h^-} = \frac{1}{R_c(T)}(T_{h^-} - T_{h^+})
$$
\n(10)

*Tablet side*:

$$
\frac{1}{R_c(T)}(T_{h^-} - T_{h^+}) = -\lambda_h \frac{\partial T}{\partial z}\Big|_{z=h^+}
$$
\n(11)

#### *3.6. The tablet/air interface and the initial conditions*

At this interface, there is no heat transfer, thus the following adiabaticity condition can be written:

$$
\frac{\partial T}{\partial z}\Big|_{z\to\infty} = 0
$$

At  $t = 0$ , the pyrotechnic composition is at thermal equilibrium with the ambient environment, so for all *z*:

$$
T(z,0) = T_{\text{ex}} = 25 \,^{\circ}\text{C} \tag{12}
$$

## *3.7. Numerical solution*

Numerical solution uses an implicit finite-difference method of order  $\Delta z^2$  for the space variable and  $\Delta t$  for time. The non-linear source term is incorporated with a  $\Delta t$  order accuracy. The time step is constant, whereas the space step follows a geometric progression so as to take account of the laser beam absorption near the surface of the tablet.

## **4.** Computation of the parameters  $R_c$  and  $\mu$

Before comparing the calculated results with the test ones, we propose methods for evaluating the two parameters which cannot be determined by test: the thermal contact resistance  $R_c$  and the absorption depth  $\delta$ . Then, the ignition characteristics are given as a function of the value of *Rc*.

#### *4.1. Evaluation of the thermal contact resistance*

A method was developed to estimate the value of *Rc* It is based on the computation of the mean equivalent air thickness, *e*, between the porthole and the tablet (Fig. 2).

Let us consider, in the first instance, that this air thickness is only due to the presence of the grains on the surface of the tablet. The grains of KClO4, assumed to be spherical, are larger than those of the reducing agent and their average diameter is about 24 µm. The air thickness is then taken as being of the order of magnitude of that of the grains, i.e., 24 µm.

The effect of porosity on the thickness is then evaluated. Given the high pressure to which powders are subjected (about 1000 bar when the pellets are pressed), it is reasonable to assume that the grains of  $KClO<sub>4</sub>$  are arranged so as to limit the space between them. The grains of iron are far smaller  $(1.6 \mu m)$ , which is why their influence is disregarded. The most efficient way to fill space with spheres is compact hexagonal stacking (known as "Kepler stacking"), and the void percentage is then 25.95%. This is particularly close to the porosity of the tablets studied, which is equal to 25.84%. It remains now to define the characteristic size, *d*, of the interstices between the spheres. An intermediate size is chosen according to Fig. 4:  $d = D(\cos(30°) - 0.5)$ , i.e.,

$$
d = 0.37D\tag{13}
$$



Fig. 4. Definition of the distance *d*.



Fig. 5. View of the first three layers, the amount of each component is respected.

where  $D$  is the diameter of particles of KClO<sub>4</sub>. Because the void proportion is 25.95% for this type of stacking, the relationship (13) is valid only for porosity values close to this value. To evaluate the equivalent air thickness contained in the tablet porosity, the percentage by mass of the composition studied:  $62\%$  iron and  $38\%$  KClO<sub>4</sub>, corresponds to a percentage by volume of 25.9% iron, 48.2% KClO4 and 25.9% air. Fig. 5 sets out the principle of the evaluation of this air thickness ( $e_{\text{poro}}$ ). Let us consider a first layer consisting of grains of KClO<sub>4</sub> and of iron surrounded by air, and let us assume that the thickness of this layer is equal to *D*. The equivalent air thickness for the next layer is obtained by multiplying by the porosity.

Thus for the layer of rank *n*:  $e_n = d\varepsilon^n$ 

In order to obtain the term  $e_{\text{poro}}$  one simply does the sum of the terms *en*. A geometric series then appears with a ratio whose value is  $\varepsilon$ . The sum of these terms is written finally:

$$
e_{\text{poro}} = d\left(\frac{\varepsilon}{1-\varepsilon}\right)
$$

In order to have the equivalent air thickness, the term *D* must be added. The equivalent air thickness *e* is written by replacing *d* by 0*.*37*d* and factoring by *d*:

$$
e = D\left(\frac{0.37\varepsilon}{1-\varepsilon} + 1\right) \tag{14}
$$

With the following data:  $D = 24 \text{ µm}$  and  $\varepsilon = 0.2584$ , the numerical application gives  $e = 27.1$  µm. This value will be used as input data for the numerical model.

# *4.2. Evaluation of the coefficient of absorption of the laser beam (Fig. 5)*

The method for determining this value is similar to the previous one: The absorption depth of each component is estimated (KClO4, iron and air) and then the absorption depth of the tablets is evaluated.

- For iron and for a wavelength of 820 nm, the absorption depth is  $\delta_{\text{Fe}} = 1.6 \times 10^{-2} \text{ }\mu\text{m}$ ;
- The KClO<sub>4</sub> is relatively transparent to the wavelength of the laser diode used. These grains are however surrounded by *iron*. The laser beam goes through the KClO4 but is absorbed by the *iron* on leaving the grain. With this assumption, the absorption depth of  $KClO<sub>4</sub>$  is of the same order of magnitude as the size of the grains. The value chosen will be  $\delta_{\text{KClO}_4} = D$ .
- The characteristic size of the pockets of air was evaluated by the relationship (14). This dimension imposes the choice of the layer thickness for the computation of the tablet's absorption depth (noted  $\delta_{\text{comp}}$ ). Given that air is transparent at the wavelength considered, for the computation of  $\delta_{\text{comp}}$  it will be necessary to take account of the successive layers of grains.
- For the tablet, the absorption length of the beam for a layer *n* can be written:

*δn* <sup>=</sup> *<sup>ε</sup>n*−<sup>1</sup>*(Xv*fer*δ*fer <sup>+</sup> *Xv*KClO4 *<sup>δ</sup>*KClO4 <sup>+</sup> *εδ*air*)*

with  $X_{\text{vfer}}, X_{\text{vKClO}_4}$  and  $\varepsilon$  the proportions by volume of each element:

$$
X_{\nu \text{fer}} + X_{\nu \text{KClO}_4} + \varepsilon = 1 \tag{15}
$$

To obtain the absorption length of the laser beam in the tablet, the sum of each layer is taken (the term  $X_{\text{vfer}}\delta_{\text{fer}}$  is negligible). After simplification, one obtains:

$$
\delta_c = D \frac{(X_{\nu \text{KClO}_4} + 0.37\varepsilon)}{1 - \varepsilon} \tag{16}
$$

For  $X_{\nu KClO_4} = 25.9\%$ ,  $\varepsilon = 25.9\%$ ,  $D = 24 \mu m$  and wavelength = 820 nm:  $\delta_c = 18.7$  µm and  $\mu_c = 5.35 \times$  $10^4$  m<sup>-1</sup>.

# **5. Results of the numerical simulations and comparison with experiments**

The computation determines the following three parameters:

 $\bullet$  the minimum pulse time  $t_{\rm pm}$  to obtain thermal ignition. Ignition is identified by a sudden runaway of the temperature in the reactive medium. The test results give a pulse time which corresponds to an initiation probability of 50% (noted  $tp_{50}$ ) associated with a dispersion  $\sigma$  if the probability density is Gaussian [10–12]. In borderline cases, when  $\sigma = 0$ , the term  $tp_{50}$  corresponds then to the minimum pulse time to obtain triggering with a probability of 100%. The result of the computation, *tpm*, can be directly compared with the test result,  $tp_{50}$ .

The minimum energy density is obtained by multiplying the value of  $tp_m$  by the power flux density (constant and verified by measurement) received by the tablet. This energy density is directly comparable to the test energy density.

- the ignition time inside the tablet (noted  $t_{ii}$ ). This time is defined by a sudden increase in the variation in the temperature (100 K for two consecutive time stages). To this time  $t_{ii}$  corresponds the tablet's ignition temperature (noted  $T_i$ ). The validity of the computation stops here. Beyond this time, phenomena, which are not taken into consideration (combustion, propagation of gases in the medium, etc.), do not enable the computation to be continued with a reliable representation of the processes involved. However, the program can continue the computation and it is then possible to define a last parameter.
- The ignition time on the tablet surface (noted  $t_{is}$ ) is always greater than the in-depth ignition time. It is defined either by a considerable gradient of the temperature of the tablet towards the outside, or by a surface temperature which is higher than *Ti*. This variable can be compared to an ignition time measured by a probe which captures the light signals emitted by the outside of the tablet. Experimentally, the time *ti* is measured between the beginning of the laser pulse and the first emissions of light [2] emitted towards the outside. The term  $t_i$  is directly comparable with  $t_i$ <sub>s</sub>.

Before presenting the simulation results, the effect of the thermal contact resistance as well as that of the absorption depth will be studied for a power flux density fixed at <sup>2</sup>*.*2 kW·cm−2.

In Fig. 6, the influence of the thermal contact resistance on the term  $tp_m$  is represented for a constant absorption depth  $(\mu = 5.35 \times 10^4 \text{ m}^{-1})$ . An increase in the thickness implies better tablet insulation with respect to the sapphire porthole. Less heat is thus given up by the tablet to the sapphire. This leads to an increase in energy efficiency as well as a reduction in sensitivity thresholds. The stabilisation of  $tp_m$  when the thickness is greater than 100  $\mu$ m shows that the tablet is completely insulated from the sapphire porthole.

Fig. 7 underlines the differences between the internal ignition time  $t_{ii}$  and surface ignition time  $t_{is}$ . It is useful to recall that the validity of the computations stops after internal ignition. However, in the case of the graph presented here, surface ignition takes place relatively shortly after in-depth ignition. It is then possible to consider that the term *tis* is a good approximation of the ignition time. It



Fig. 6. Effect of the insulator thickness e on pulse time *tpm*.



Fig. 8. Ignition depth of pellet.

is clearly established that a lesser air thickness implies a greater difference between the values of *tii* and *tis*. Because the time  $t_{ii}$  corresponds to the internal ignition and  $t_{is}$  to the surface ignition, the difference  $t_{is} - t_{ii}$  is, in fact, the time corresponding to the distance covered by the reaction front. It is then possible to compute the average rate of advance of the reaction front. For a given pyrotechnic mixture, one can assume that the more significant this difference, the more ignition takes place at depth. This assumption is verified in Fig. 8.

The difference  $t_{is} - t_{ii}$  is obtained for various insulator thicknesses (Fig. 8). The smaller the thickness, the more ignition takes place at depth. On the other hand, if the thickness exceeds 100 µm, ignition occurs closer to the surface  $(t_{is}$  is always higher than  $t_{ii}$ ). This phenomenon is explained physically by the presence of the porthole which absorbs the heat coming from the surface of the tablet with greater or lesser efficiency according to the value of the air thickness, *e*. When the sapphire absorbs a lot of heat (that is for small *e*), the temperature maximum shifts towards the



Fig. 9. Temperature profiles at  $P = 0.6$  W: (a) Insulator thickness: 36 µm,  $tp_m = 1.77$ ,  $t_{ii} = 1.84$ ; (b) Insulator thickness: 0.4 µm,  $tp_m = 4.39$ ,  $t_{ii} = 4.52$ .



Fig. 10. Differences between temperatures  $T_{\text{max}}$  and  $T_h^+$  of pellet: (a)  $e = 36 \text{ µm}$ ; (b)  $e = 0.4 \text{ µm}$ .

inside of the tablet until there is equilibrium between the heat losses by conduction, before and after the direction of the laser beam. This equilibrium is reached for a depth which varies from a few microns to about twenty microns.

In Fig. 9(a) and (b), two temperature against depth, *z*, profiles are represented for two extreme values of the air thickness ( $e = 36$  µm and  $e = 0.4$  µm), for various times. The depth corresponding to  $z = 1$  mm coincides with the interface between the tablet and the porthole. The temperature profiles are represented until the internal ignition is obtained. A general observation of these curves confirms the previous assumptions:

- on the interface (porthole side), the temperature is greater if  $e = 0.4$  µm, which corroborates the fact that the porthole absorbs more heat when the air thickness is less.
- in Fig. 9(b) when  $t = 4.4$  ms, the maximum temperature in the porthole is about  $80^{\circ}$ C and this temperature variation is far smaller than that of the pyrotechnic medium. This justifies the use of a constant step meshing for this field.
- in the case where the insulation is greatest  $(e = 36 \text{ µm})$ , the maximum temperature difference at the interface does not exceed 550 K. This justifies the fact that we disregarded the radiation at this interface.
- for a small air thickness, the ignition occurs at depth. In Fig. 9(b), ignition is situated 17.2 µm behind the porthole.

The steepest temperature gradients are very close to the interface with the pyrotechnic mixture. Fig. 9(a) and (b) show the very large temperature variation in the pyrotechnic composition.

In Fig. 10, the differences between the maximum temperature  $(T_{\text{max}})$  and the surface temperature  $(T_h^+)$  of the pyrotechnic medium are represented. From the very first moments of laser heating, the difference increases uninterruptedly. It proves that the hottest point shifts towards the inside of the tablet. The differences increase significantly when the insulator thickness increases.

Fig. 11 shows the variation of the depth corresponding to the maximum temperature as a function of time. It confirms that the temperature maximum is at an increasing depth as the time increases. From these graphs one can connect the time corresponding to the internal ignition  $t_{ii}$  to the depth *z*maximum:

$$
z_{\text{max}} = at^b \tag{17}
$$

The constants *a* and *b* depend on the insulator thickness *e*. All of the computations showed that this law is verified irrespective of the thickness *e* and this with very good accuracy. The relationship (17) makes it possible to deduce directly the propagation rate of the reaction front in the tablet which is defined by:  $v = \frac{\partial z_{\text{max}}}{\partial t} = abt^{b-1}$ . Given that *b* is always less than 1, the rate always decreases until the ignition identified by  $t_{ii}$ . Fig. 12(a) and (b) illustrate the stabilisation of the depth  $z_{\text{max}}$  which precedes ignition.

The computed results are compared in Fig. 13 with the test results obtained at the LEES laboratory. The thickness  $e$  and the coefficient of absorption  $\mu$  previously computed



Fig. 11. Depth where temperature  $T_{\text{max}}$  takes place: (a)  $e = 36 \text{ µm}$ ; (b)  $e = 0.4 \text{ µm}$ .



Fig. 12. Speed of  $T_{\text{max}}$  displacement inside the pellet and before ignition: (a)  $e = 36 \text{ µm}$ ; (b)  $e = 0.4 \text{ µm}$ .

have been used as model's input data. The ignition thresholds obtained by test and computation are close and the difference is of the order of 20%. However, for the lowest power flux density *(*0*.*75 kW·cm−2*)*, the correspondence is less precise. The methods of computation of the parameters  $e$  and  $\mu$  thus seem sufficient to account for the test trends. These parameters are highly sensitive because little variations in them could change the thresholds by a factor of three.

Taking the porthole into account makes it possible to consider the heat losses. However, this cannot, on its own explain, the phenomenology of the energy exchange. If there is no contact resistance, the thresholds are far too great. Taking a thin gas thickness into account is of the utmost importance in order to limit the losses. This thickness is naturally included in the tablet's surface roughness which itself depends on the porosity and surface condition of the piston which was used for the moulding. In Figs. 6 and 7, one can note that, if the air thickness is too small (with  $\mu = 5.35 \times 10^{-4} \text{ m}^{-1}$ ), ignition becomes impossible. In fact, when *e* tends towards zero, one finds exactly the case where the layer of air is not taken into consideration. The energy supplied by the laser diode is partly spread in the porthole thus preventing a sufficient concentration of energy to cause the ignition of the tablet.

After having examined the influence of the parameters *e* and  $\mu$  on the ignition energy thresholds, we compare the results obtained with the Opdebeck tests [7]. Fig. 13 shows that the computed ignition thresholds correspond with the test results. When the power flow density is greater than <sup>1</sup>*.*5 kW·cm−2, the numerical results are very close to the



Fig. 13. Effect of power density on ignition thresholds.

tests. However, they remain slightly less than the measured thresholds. Other computations, which corroborate the results in Fig. 13, made it possible to study the variation of the energy density thresholds as a function of the power flux density, and for several thicknesses *e*. They showed that a reduction in *e* necessarily implies a general increase in energy threshold. That means that the air thickness is overestimated for this model.

## **6. Conclusion**

The results of the computations show that the contact with the porthole used in the test leads to a very significant loss of energy by absorbing the heat released in the pyrotechnic tablet. Consequently, the ignition energies increase. The computed ignition thresholds are closely linked to the value given to this thickness: too small a thickness can ruin any ignition attempt. The computations carried out in connection with this model have supplied both practical and fundamental information. It was clearly established that a smaller air thickness leads to a greater difference between the moment of internal ignition  $t_{ii}$  and external ignition  $t_{is}$ . The delay  $t_{is} - t_{ii}$  is identified with the time that the reaction front takes to move from the inside of the tablet towards the surface, which makes it possible to define the average rate of the reaction front. The study proved that the depth at which ignition occurs is related to this delay. A longer time, as well as a significant insulator thickness, causes ignition at greater depth.

For each ignition, a characteristic common to all the combinations of the value  $(\mu, e)$  emerges. Ignition is always preceded by a stabilisation of *z*max. It thus seems that this stabilisation is a necessary condition for ignition. In other words, before the energy released by chemical reactions increases in power, it is necessary for there to be an equilibrium between the thermal diffusion from the hottest point on the tablet towards the porthole and the diffusion from the hottest point towards the inside of the tablet.

By introducing actual contact, a new variable has to be determined: the thermal contact resistance. A computation method to estimate the gas thickness has been proposed. A computation method makes it possible to obtain the depth  $\mu$  of absorption in the tablet, for the laser wavelength used (820 nm). The values of  $\mu$  and of  $e$  are used so as to compute the energy thresholds under the same conditions as the test. The computations thus obtained are very much in keeping with the tests. The methods to estimate the values of the coefficient of absorption  $\mu$  and the insulator thickness  $e$  are therefore satisfactory. However, the difference between tests and computation is rather significant when one uses low powers.

The results obtained by this simulation make it possible to improve the understanding of the phenomena which govern ignition for test conditions. This study advances the very great dependence of the energy thresholds as a function of the value of *e*.

An increase in the insulator thickness *e* leads to better insulation of the tablet, which makes it possible to reduce the thermal sensitivity thresholds. This behaviour makes it possible to draw a conclusion according to the test mode chosen for ignition:

• by devising an ignition system which uses a re-focussing of the laser beam, it is necessary to envisage a greater distance between the pyrotechnic composition and the porthole which protects the optical system. If one refers to the computations, the thermal behaviour of the window/tablet system no longer changes for an insulator thickness greater than 100 µm. In order for the beam to converge on the tablet and not on the face of the porthole, it is essential to take account of this distance in order to concentrate the laser beam on the surface of the tablet.

• During the design of a system of ignition by direct contact of an optical fibre, it is necessary to envisage an optimum distance between the pyrotechnic composition and the fibre. Indeed, as was shown previously, it is necessary to provide a sufficient distance in order to avoid the absorption of the heat by the optical fibre (particularly if it is made up of a somewhat conductive material like silica), or to avoid damaging it (polymer fibre). But unlike the previous system, imposing a significant distance (between the face of the optical fibre and the tablet surface) leads to a great disadvantage. In this case, if the distance is too long, the diameter of the laser spot will be bigger on account of the property of divergence of the beam as it leaves the fibre. The power flux density available will be smaller, which will not favour ignition.

In the same manner as for this study, it would be interesting to try to find the variation in the parameters  $E_{50}$  and  $t_i$  as a function of the value of the coefficient of absorption. Further fundamental conclusions could be supplied, as well as practical information for the design of a new ignition system.

## **References**

- [1] Ph. Gillard, M. Roux, Ignition of pyrotechnic mixture by means of a laser diode, Part I: Numerical modelling, Propellants Explosives Pyrotechnics 22 (1997) 256–262.
- [2] E. Radenac, Ph. Gillard, M. Roux, Laser Diode ignition of the combustion of pyrotechnic mixtures. Experimental study of the ignition of Zr/KClO4 and Zr/PbCrO4, in: 29th International Annual Conference of ICT, 1997, pp. 40.1–40.14.
- [3] E. Radenac, Numerical and test study of the ignition of pyrotechnic compositions by a laser diode, University of Poitiers thesis, 1998.
- [4] J.S. Lee, L.K. Lin, C.H. Lin, J.P. Chien, C.W. Huang, S.S. Chang, A study of zirconium/potassium perchlorate primer mixtures, Thermochim. Acta 173 (1990) 211–218.
- [5] C. Brassy, F. Marlin, M. Roux, Ignition mechanism of the Zr/PbCrO4 pyrotechnic mixture in presence of an oxidising atmosphere, Propellants Explosives Pyrotechnics 19 (1994) 165–170.
- [6] E. Radenac, F. Faubert, Ph. Gillard, Influence of the particle size distribution of iron on the threshold of initiation of the laser heating of the Fe/KClO4 mixture, Internat. Pyrotechnics Congress EURO PYRO 99 (1999) 528–539.
- [7] F. Opdebeck, E. Radenac, P. Gillard, Laser diode ignition of the Iron/KClO4 pyrotechnic mixture: Experimental and parametrical study, in: 32nd international annual conference of ICT, 2001, pp. 3.1–3.12.
- [8] B.L. Hicks, Theory of ignition considered as a thermal reaction, J. Chem. Phys. 22 (1954) 414–429.
- [9] R.P. Tye, Thermal Conductivity, Vol. 1, Academic Press, New York, 1969.
- [10] Statistical method of measurement. Bruceton Test, Space pyrotechnics working party, GTPS n◦11C, 1982, pp. 1–11.
- [11] Statistical method. Dichotomic approach, Space pyrotechnics working party, GTPS n◦11E, 1993.
- [12] H.J. Langlie, A reliability test method for "one shot" item, Aeronautronic, Newport Beach, CA, Report U-1792, 1962.