

# Numerical simulations of ignition and combustion of RDX mixed with gaseous additives

Modou Mar, Philippe Gillard, Léo Courty  
Univ. Orléans, PRISME EA 4229, Bourges, France

## 1 Introduction

The use of energetic materials in propulsion field leads to several safety problems. Indeed, accidental stresses from thermal (fires) or mechanical (storage, transport) point of view can be of importance when it comes to ensure efficiency in spatial technologies. Today, very sensitive propellants are mainly used for propulsion. These powders can therefore lead to accidental situations and create important damages to the amenities and to human lives. Important accidents are those of the USS Oriskany (1966) and of the USS Enterprise (1696). To reduce the risk of accidental ignition, low sensitivity propellants have been developed in the early 90's. These powders allow better conditions of transport and storage than the "classical" ones. Pure RDX (1,3,5-trinitrohexahydro-s-triazine) is used in the composition of several low sensitivity energetic materials. There are few data in the literature on the combustion of RDX. The aim of this work is to study the effects of different parameters on the combustion of RDX. Different simulations with several initial mass, temperature, pressure, additives and heat transfer coefficient are performed with COMSOL software using a detailed chemical kinetic mechanism. A literature survey has been done on the problem of ignition of propellants for different initial pressures and we found that this parameter and the nature of gaseous additive are of importance. During the past decades, many theoretical complex models for RDX combustion have been developed. Early works were conducted in 1966 by Price et al. [1] and in 1982 by Kulkarni et al. [2]. A detailed reaction scheme of RDX steady-state combustion was formulated by Melius [3], considering the condensed-phase region as a dense gas. The kinetic scheme developed by Melius was later improved by Yetter et al. [4] and used in steady-state combustion modeling by Prasad et al. [5]. Liau and Yang [6] recognized that the condensed phase was as an important aspect of RDX ignition by accounting for a foam layer (that consisted in liquefied RDX, bubbles of gaseous RDX and its decomposition products), in which a global decomposition mechanism of RDX was considered. Our simulations were performed using the detailed chemical kinetic mechanism proposed by Yetter et al. [4]. This mechanism, COMSOL Multiphysics® Modeling Software and the studied parameters are briefly described in the next section. Section 3 is devoted to the presentation of results and discussions of final temperature, pressure, maximum rate of explosion pressure rise and species concentrations as functions of time.

## 2 Theoretical formulation

In this work, numerical simulations for initial masses ranging between 0.3 and 1 g (with a 0.1 step) were performed. Initial pressures and temperatures were studied respectively between 1 and 10 bars and between 300 and 1100 K. Different heat transfer coefficients were studied in order to show the influence of convection and radiation near the wall. The detailed chemical kinetic mechanism is the one proposed by Yetter et al. [4]. This mechanism is composed of 231 elementary reactions and 45 species. The condensed phase is not included in this study and the gas phase is composed of equivalent RDX vapors, additives and reaction products. We were interested in final temperature, pressure, maximum rate of explosion pressure rise and species concentrations as functions of time. Simulations were carried out thanks to COMSOL Multiphysics® Modeling Software by solving detail kinetic mechanisms, conservation equations of mass, energy and species concentrations. The ignition model of RDX is spatially 0D and does not include the transient of the condensed phase into gas phase. The condensed-phase region was treated as a dense gas. The gas phase is composed initially of RDX vapor. It is assumed that the gas mixture is composed of gases obeying the ideal gas law. Thus, the ideal gas law for multi-species is derived to close the formulation:

$$p = R_g T \sum c_i \quad (1)$$

where  $c_i$  is the species molar concentration,  $T$  is the temperature, and  $R_g$  is the ideal gas constant.

The species mass balances are:

$$\frac{dc_i}{dt} = R_i \quad (2)$$

where  $R_i$  is the species rate expression.

For an ideal reacting gas, the energy balance is:

$$V_r \sum_i c_i C_{p,i} \frac{dT}{dt} = Q + Q_{ext} - V_r \frac{dp}{dt} \quad (3)$$

where  $V_r$  denotes the reactor volume,  $C_{p,i}$  is the species molar heat capacity and  $p$  is the pressure. On the right hand side  $Q$  is the heat due to chemical reaction, and  $Q_{ext}$  denotes heat added to the system. The heat of reaction is:

$$Q = -V_r \sum_j H_j r_j \quad (4)$$

Where  $H_j$  is the enthalpy of reaction, and  $r_j$  the reaction rate.

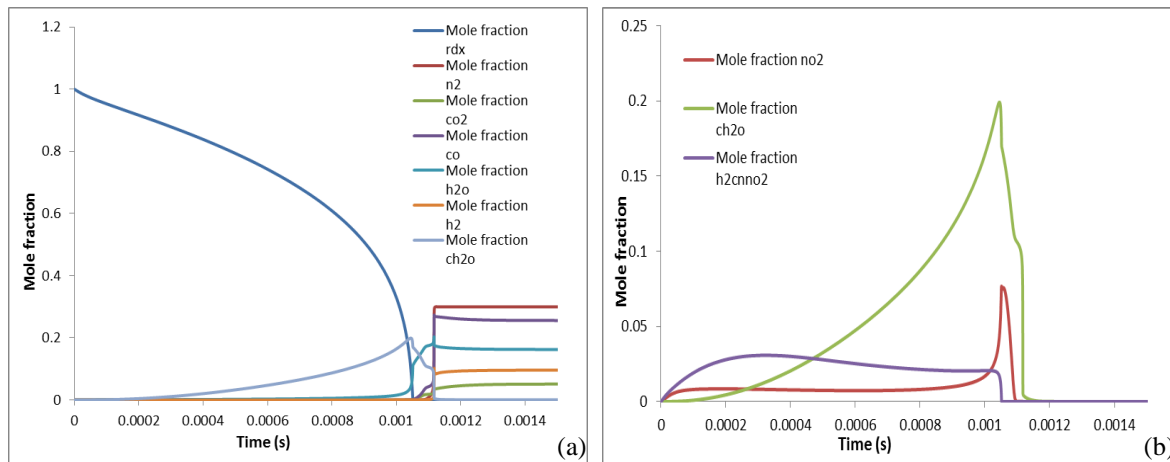


Figure 1. Species mole fraction evolution as a function of time during RDX combustion for initial pressure and temperature of 1 bar and 700 K.

### 3 Results and discussions

#### Species evolution

An example of temporal evolution of species concentration during the combustion process is shown in figure 1. Here, the first step in the gas-phase decomposition of RDX indicates that the initial species produced are mainly  $\text{NO}_2$ ,  $\text{H}_2\text{CNNO}_2$  and  $\text{CH}_2\text{O}$  (figure 1 (b)). These species are consumed quickly at high temperature and allow the formation of final products, including  $\text{N}_2$ ,  $\text{CO}_2$ ,  $\text{CO}$ ,  $\text{H}_2\text{O}$ ,  $\text{H}_2$ ,  $\text{NO}$  and  $\text{CO}_2$  among others. Results with other initial pressures and temperatures show a similar trend.

### Effect of initial temperature

For all results, ignition occurs only if the initial temperature is greater than or equals to 600 K. Figure 2 gives an example of the temporal evolution of temperature during the combustion of pure RDX vapors for two different initial temperatures (600 K and 1100 K). We note that the gaseous RDX ignition process become shorter when increasing initial temperature. We can also note that the temperature profile depends on the species evolution, in particular that of  $\text{H}_2\text{O}$ .

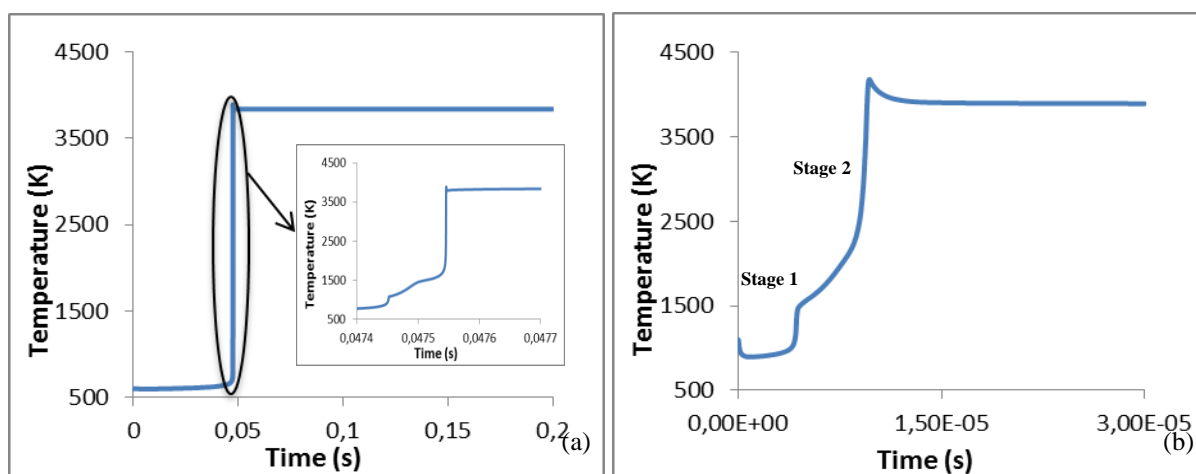


Figure 2. Temperature as a function of time for the combustion of RDX: (a) initial temperature of 600 K and (b) initial temperature of 1100 K.

The ignition process of gaseous RDX is clearly divided into two distinct stages (figure 2): first, we have the RDX decomposition process that depends on the initial temperature. In Stage 2, we have oxidation reactions, mainly caused by the first species produced, followed by highly exothermic reactions and the formation of all final products. After this stage, no reaction occurs.

Table 1. Pressure, temperature and ignition delay of RDX combustion for several initial temperatures.

	Initial Temperature (K)					
	600	700	800	900	1000	1100
<b>Pressure (bar)</b>	62	54	47	43	39	36
<b>Temperature (K)</b>	3840	3848	3858	3869	3875	3892
<b>Ignition delay (ms)</b>	45	1	1,60E-01	5,67E-02	2,30E-02	1,90E-03

Table 1 lists the predicted results of maximal pressures, temperatures and ignition delays (defined as the time required for the temperature to reach the maximal temperature from the initial temperature). The values of ignition delay are small compared with those obtained by Liao and Yang [6]. This difference is due to the fact that, in our study, the overall gaseous RDX ignition process does not take into account completion stages of the RDX decomposition process and solid-gas interface phenomena.

### Effect of initial pressure

The dependence of the initial pressure on final temperature, pressure, species concentrations and ignition delay is shown in figure 3. We note that the final pressure increases proportionally to the initial pressure. Final pressure is much more dependent from initial pressure than final temperature. The ignition delay becomes shorter and decreases slower when the initial pressure is increasing. This RDX behavior is similar to the one of other hydrocarbons studied recently in the literature, like methane [7] or n-decane [8]. We can note the case of nitromethane, where initial pressure seems not to have any effect on ignition delay times [9].

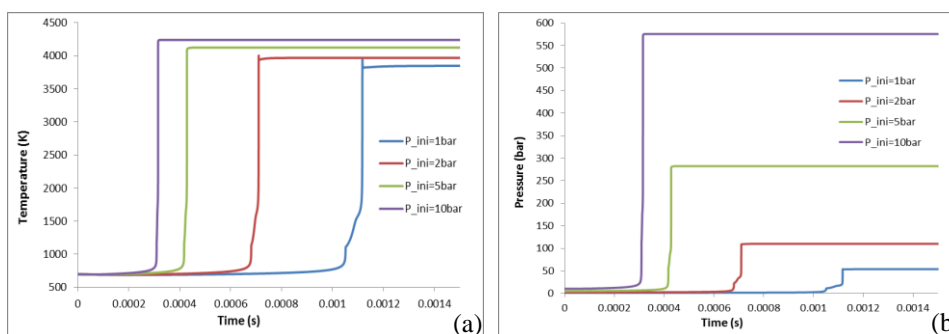


Figure 3. Temperature (a) and pressure (b) as a function of time for different initial pressures for the combustion of RDX for an initial temperature of 700 K.

#### Effect of initial mass

The results show that initial mass of RDX does not influence the final temperature (figure 4), but only final pressure. In fact, increasing the mass of RDX generates higher concentrations of gaseous species involved in the ideal gas law.

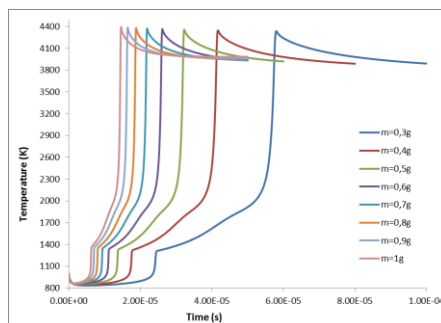


Figure 4. Temperature as a function of time for different powder initial masses for the combustion of RDX for initial temperature and pressure of 1000 K and 1 bar.

#### Effect of additives addition

Figures 5 and 6 show the influence of additives addition on the final temperature and pressure respectively. Ignition delays are increasing with the addition of Ar, N<sub>2</sub>, Air or O<sub>2</sub>. The main increase is observed with the addition of N<sub>2</sub> that cannot be considered as an inert gas for RDX combustion. Lower final temperature and pressure are also obtained with the addition of this species.

The current model shows that the addition of Ar, N<sub>2</sub>, Air or O<sub>2</sub> controls the final pressure and the ignition process of a mixture containing 90% of RDX. Ostmark and Gräns [10] performed experiments in air and N<sub>2</sub> of RDX combustion and found that ignition delays were strongly increased in nitrogen. This is in agreement with our simulation results.

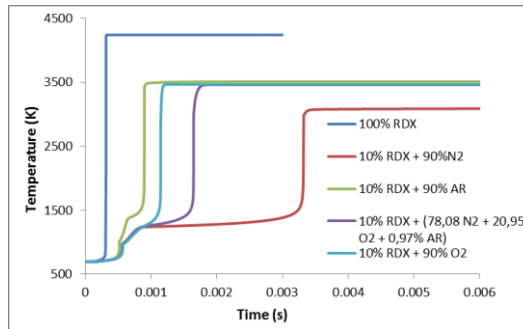


Figure 5. Temperature as a function of time for the combustion of RDX mixed with different additives for initial temperature and pressure of 700 K and 10 bar.

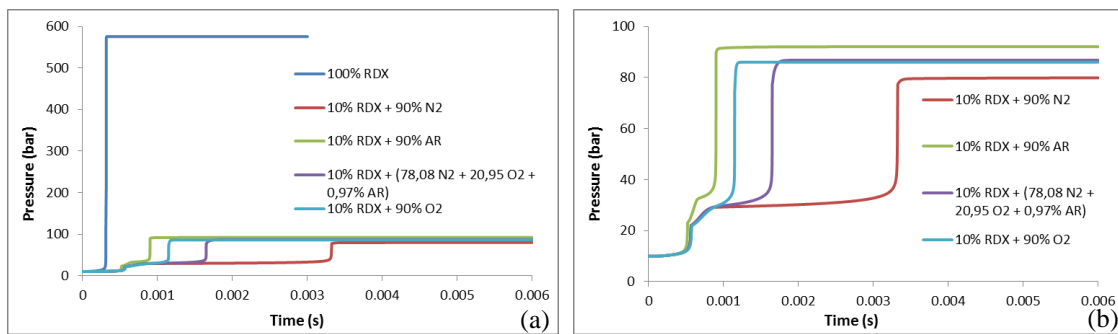


Figure 6. Pressure as a function of time for the combustion of RDX mixed with different additives for initial temperature and pressure of 700 K and 10 bar (a); zoom on the case with additives for the same conditions (b).

The initial temperature necessary to ignite the mixture and the ignition delay can considerably reduce by adding OH radicals (figure 7). Without these radicals, it not possible to ignite under 600 K at atmospheric pressure. Similar behavior is generally observed for hydrocarbons.

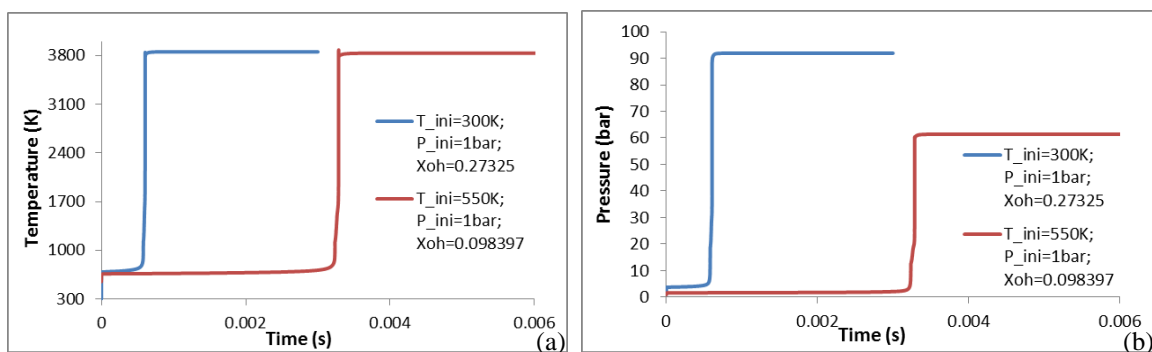


Figure 7. Temperature (a) and pressure (b) as a function of time for the combustion of RDX mixed with OH radicals for several initial temperatures and an initial pressure of 1 bar.

**Effect of heat transfer coefficient**

Simulations are conducted taking into account convective heat transfer coefficient ( $h$ ) from 0 to 1 W/cm<sup>2</sup>.s. Results are presented in figure 8. We note that a high value of  $h$  is needed to see the effect of heat loss at the walls.

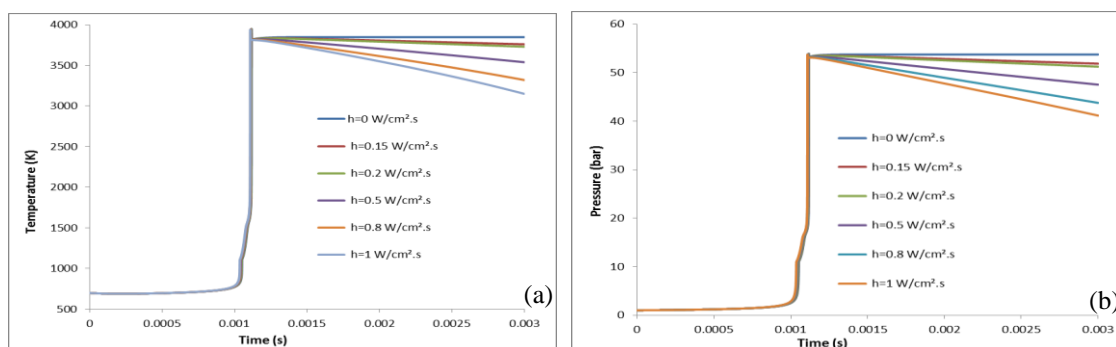


Figure 8. Temperature (a) and pressure (b) as a function of time for the combustion of pure RDX with different heat transfer coefficients.

## 4 Conclusion

Numerical simulations were performed to provide an understanding of the ignition processes of RDX. A detailed kinetic mechanism was used for the gas phase. The effects of initial temperature, initial pressure, gas addition and heat transfer coefficient were studied. Below 600 K, the reaction may take place with the addition of OH radicals. Temperature and pressure are drastically reduced with the addition of nitrogen. On the contrary, ignition delays are magnified. The addition of argon or air also increase ignition delays compared to pure RDX. Overall, the model is capable of providing reasonable prediction of ignition of gaseous RDX. This study belongs to a global research project called Lilovem that plan to determine the influence of initial pressure of some gases in a laser ignition bench.

**Acknowledgment:** *Authors want to thank the ‘Conseil Régional du Centre’, in France, which has financially supported this work.*

## References

- [1] E.W. Price, H. H. Bradley Jr., G. L. Dehority, M. M. Ibricu, Theory of Ignition of Solid Propellants, AIAA J., 4, 1966, 1153-1181.
- [2] A. K. Kulkarni, M. Kumar, K. K. Kuo, Review of Solid-Propellant Ignition Studies, AIAA J. 1982, 20, 243-244.
- [3] C.F. Melius, Thermochemical Modelling II, Application to Ignition and Combustion of Energetic Materials, in Chemistry and Physics of Energetic Materials, Bulusu ed., Vol. NATO ASI 309, Kluwer Academic, Norwell, MA, 1990, 21-78.
- [4] R.A. Yetter, F.L. Dryer, M.T. Allen, J.L. Gatto, Development of Gas-phase Reaction Mechanisms for Nitramine Combustion, Journal of Propulsion and Power, 4, 1995, 683-697.
- [5] K. Prasad, R.A. Yetter, M.D. Smooke, An eigenvalue method for computing the burning rates of RDX propellants, Combustion Science and Technology, 124 (1), 1997, 35-82.
- [6] Y.C. Liao, V. Yang, Analysis of RDX monopropellant combustion with two phase subsurface reactions, Journal of Propulsion and Power, 11 (4), 1995, 729-739.
- [7] C. Tang, L. Wei, J. Zhang, X. Man, Z. Huang, Shock tube measurements and kinetic investigation on the ignition delay times of methane/dimethyl ether mixtures, Energy&Fuels, 26, 2012, 6720-6728.
- [8] X. Hui, C. Zhang, M. Xia, C.J. Sung, Effects of hydrogen addition on combustion characteristics of n-decane/air mixtures, Combustion and Flame, 161, 2014, 2252-2262.
- [9] A. Ambekar, R. Bhangale, R. Chatterjee, C. Kulkarni, S. Kumar, A. Chowdhury, Glow-plug-assisted combustion of nitromethane sprays in a constant volume chamber, Applied Thermal Engineering, 76, 2015, 462-474.
- [10] H. Östmark, R. Gräns, Laser ignition of explosives : effects of gas pressure on the threshold ignition energy, Journal of Energetic Materials, 8 (4), 1990, 308-322.