Laser ignition of a low-vulnerability RDX-based propellant: influence of the atmosphere on ignition and combustion properties

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1 Introduction

Propellants are energetic materials especially employed for space rocket propulsion or weapons. Historically, nitrocellulose was widely used for the formulation of solid smokeless propellants like single or double-base propellants. Nevertheless, the quest for safer propellants led to the decrease of the nitrocellulose content for more stable ingredients. For this purpose, RDX represents an excellent candidate which also enables high specific impulse along with less smoke, toxicity and corrosion.
Combustion of solid propellants is a complex multiphase phenomenon involving numerous processes such as thermal decomposition along with subsequent reactions coupled with molecular diffusion, convection, conduction and radiation. So as to improve our comprehension of these processes, numerous studies were performed, especially regarding combustion of RDX-based propellants.
The modelling of RDX combustion has considerably flourished at the end of the last century. With the development of computing resources, detailed kinetics chemistry including thermal decomposition for the gas phase could be implemented. In 1990, Melius formulated a detailed scheme for RDX reaction mechanism including 38 species and 158 reactions [1]. Yetter et al. [2] then improved this scheme which was used by Prasad et al. [3] for the modelling of steady-state combustion of RDX. These schemes have been the basis of ensuing research work. Liau et al. first accounted for the presence of a foam layer, made of liquid and gas bubbles, in which a global decomposition mechanism derived from Brill et al. was considered. The foam layer was mathematically represented by a void fraction in the condensed-phase equations [4]. This formalism was further extended to the gas phase by considering the presence of condensed particles. It was detailed by Beckstead et al. in their review of modelling of solid propellants combustion and ignition in 2007 [5]. More recently, Patidar et al. developed a detailed mechanism for the liquid-phase decomposition of RDX and subsequent reactions [6]. They proposed new decomposition pathways which were included in the Chakraborty's scheme for the gas phase [7].
The huge variety of energetic materials would require an infinity of experiments to be precisely characterized. From this perspective, modelling enables reduction in time consumption, cost and improvement of safety. However, it is not yet completely predictive and experimental work is still necessary. The aim of this work is to study the influence of atmosphere on ignition and combustion properties of a low-vulnerability RDX-based propellant ignited by a laser diode. A previous study, employing this material, was published by Gillard et al. comparing results for nitrogen and argon

atmospheres [8]. Here, experiments were conducted under synthetic air. Few data are available on ignition behavior and combustion characteristics of this kind of propellant. On the contrary, studies seem rather incline towards more sensitive RDX materials. Fang et al. recently demonstrated that incorporation of gold nanoparticles in RDX crystals could reduce thresholds by an order of 3 compared with pure RDX [9]. In 2018, Aduev et al. reported similar results with RDX containing ultrafine aluminum particles [10]. The next section details the experimental aspects of the study. Section 3 is dedicated to results and discussions.

2 Experimental details

RDX-based propellant

The material of interest is a low-vulnerability gun propellant composed of 84 % of RDX and 16 % of a binder, mainly HTPB. These pellets are commercial propellants provided by ArianeGroup. They do not suffer any treatment except heating during 24 h at 50 °C to remove discrepancies due to humidity before tests. As presented in Figure 1, pellets are perforated cylinders and contain 7 holes. The average mass, diameter and length are respectively 26 mg, 2.7 mm and 3.8 mm.

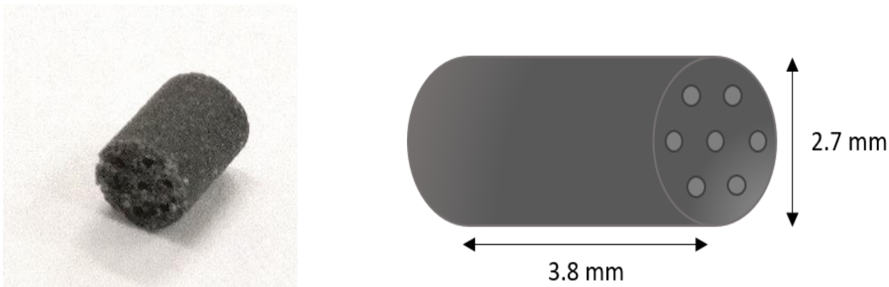


Figure 1: Picture and scheme of the propellant pellet

Experimental setup

Pellets are ignited in a stainless steel cylindrical closed-volume vessel of 55 cm^3 by the means of a laser diode (Coherent FAP System, λ = 808 nm) coupled to an optical system and a pressure sensor. An overview of the setup is presented Figure 2. At the end of the laser diode, an optical fiber brings the laser beam to the optical system composed of two plano-convex spherical lenses with focal lengths of 16 and 25 mm. The latter enables to obtain a spot diameter of 1.25 mm on the sample. A pressure sensor (Kistler 603 B) is connected to a digital oscilloscope (Tektronix DPO2014B).

An experiment is conducted as follows: a pellet is placed in a PMMA holder placed itself in the combustion chamber and closed by a sapphire window. Samples are placed against the window. Reactor is then pressurized with argon, nitrogen or synthetic air (20 % O_2 + 80 % N_2) at a pressure ranging from 10 to 60 bar. Finally, laser energy is brought and pressure signal is recorded if ignition occurs. Laser pulse and power can be varied and laser temperature is fixed at 20 °C. Five powers were studied: 1.43, 2.86, 5.00, 6.42 and 9.95 W which correspond to laser current intensity of 10, 12, 15, 17 and 22 A. For each power, the delivery of energy is controlled by the duration of the laser pulse (t_{pulse}).

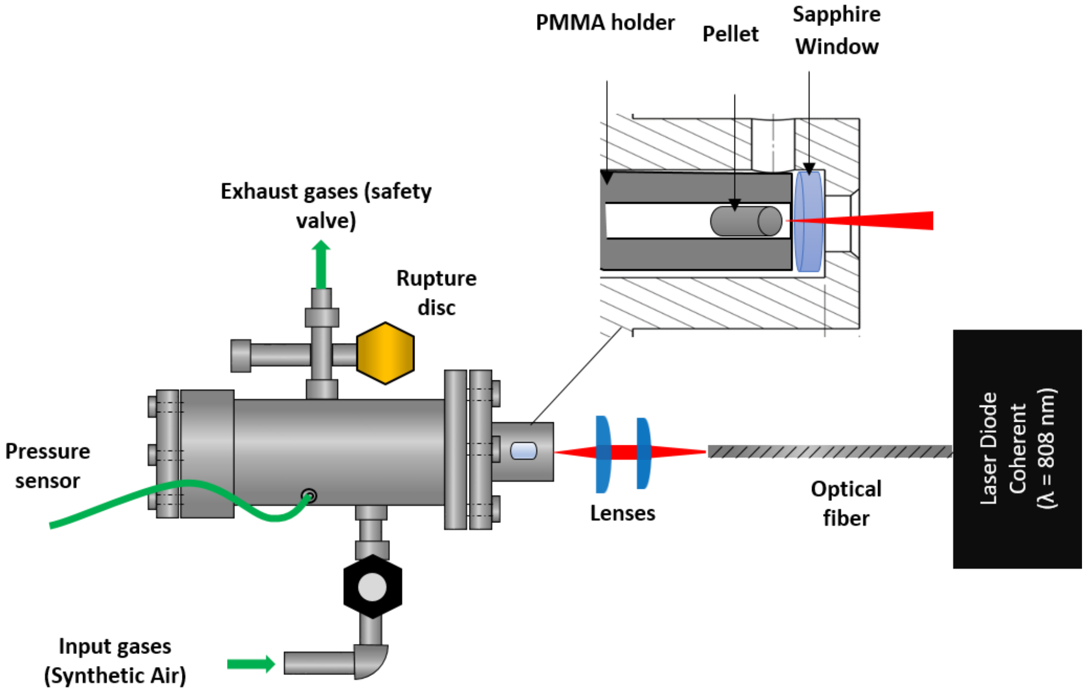


Figure 2: Scheme of the experimental setup

3 Results and discussions

Overpressures (ΔP), ignition delays (t_i) and propagation rates (r_P) are derived from the pressure signal. t_i is defined as the time between the beginning of the laser pulse and the beginning of the increase in pressure. ΔP represents the maximum value of the pressure signal. It is divided by the mass of the pellet (m) to make results independent of this parameter. Regarding r_P it is computed as the maximum value of the derivative of the pressure signal divided by mass $\frac{1}{m}\left(\frac{d(\Delta P)}{dt}\right)_{max}$.
Minimum ignition energies (E_{50}) are determined by Langlie method. For a given atmosphere, initial pressure (P_0) and laser power (P_{laser}), E_{50} represent energies giving 50 % of probability of ignition. Langlie method is a statistical method based on dichotomy principle which assumes a repartition of ignition threshold following a standard normal distribution [11].
Results on ΔP, t_i and r_P are presented as functions of three parameters: atmosphere (air, argon, nitrogen), initial pressure and laser power. E_{50} are presented as functions of atmosphere and laser power at P_0 = 50 bar. This section is divided into two parts: influence of initial pressure and atmosphere and influence of laser power and atmosphere.

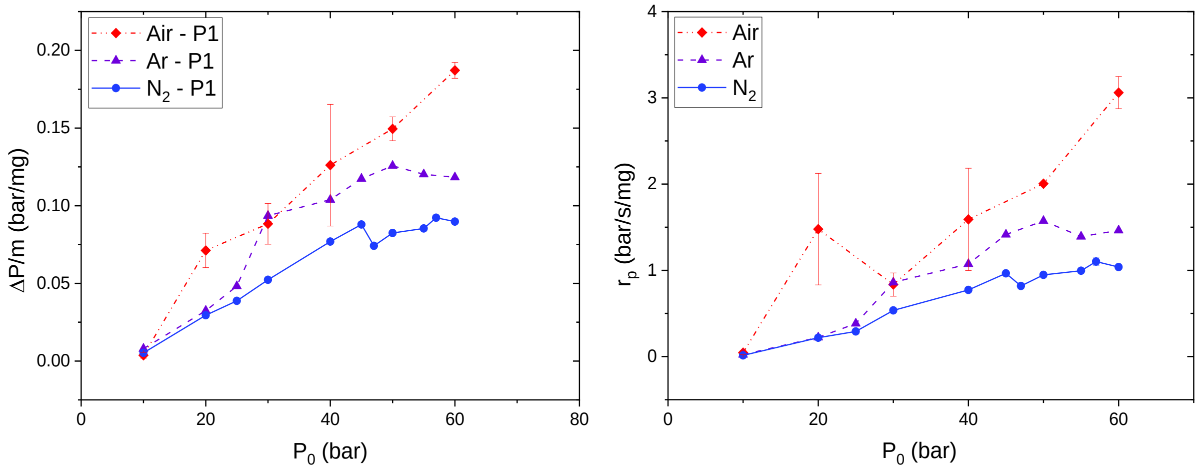
Influence of initial pressure and atmosphere

These results have already been presented in [8] for argon and nitrogen, here, comparisons are made with synthetic air. Figure 3 presents overpressures (a) and propagation rates (b) as functions of initial pressure for the three different atmospheres and at P_{laser} = 2.86 W. As already noted, overpressures and propagation rates are globally increasing with initial pressure. However, there are some critical pressures for which this trend is not observed: at 30 bar for air, 47 bar for nitrogen and 55 bar for argon. With reference to atmosphere, it is clear that overpressures and propagation rates are higher under air except at 10 and 30 bar. This observation could find an explanation in the fact that oxygen balance of RDX is negative: OB = -21.6%. Indeed, oxygen present in RDX molecule is not sufficient to completely oxidize the other components. Under air atmosphere, exothermic reactions such as (1) or (2) could be enhanced, releasing then more heat and leading to greater temperatures and pressures:

$$2CO + O_2 \leftrightarrow 2CO_2 \qquad (1)$$

$$2NO + O_2 \leftrightarrow 2NO_2 \qquad (2)$$

Hiyoshi and Brill have reported the great importance of reaction (1) in RDX/air combustion (~70 % of CO_2 and 0 % of CO in products) but do not really enable to conclude on its enhancement compared with argon. Reaction (2) as for it seems to have a minimal role in the combustion process since little NO and NO_2 were detected after ignition [12]. In previous studies, the best combustion properties under argon, compared with nitrogen, were explained by a lower molar heat capacity at constant volume enabling to reach higher temperatures and pressures [8].



(a) *(b)*

Figure 3: Overpressures and propagation rates as functions of initial pressure and atmosphere, P_{laser} = 2.86 W, t_{pulse} = 500 ms

Ignition delays against initial pressure at P_{laser} = 2.86 W are plotted Figure 4. First, we can notice ignition delays tend to decrease with increasing initial pressures. Moreover, there is no clear difference between shots under argon and nitrogen. At P_0 = 20 bar, ignition delays under air are slightly better than others. The differences air/argon and air/nitrogen seem to be maximal at P_0 = 40 bar before decreasing again.

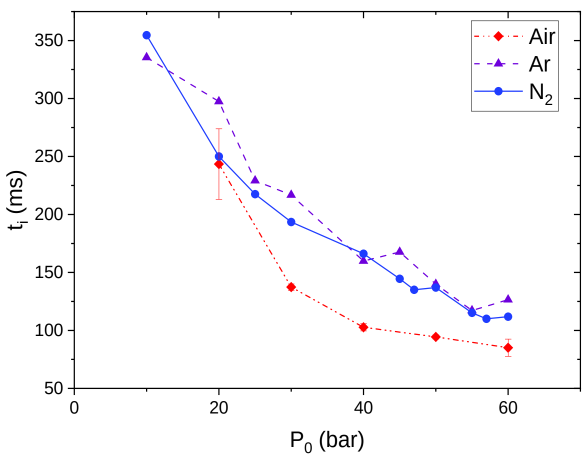
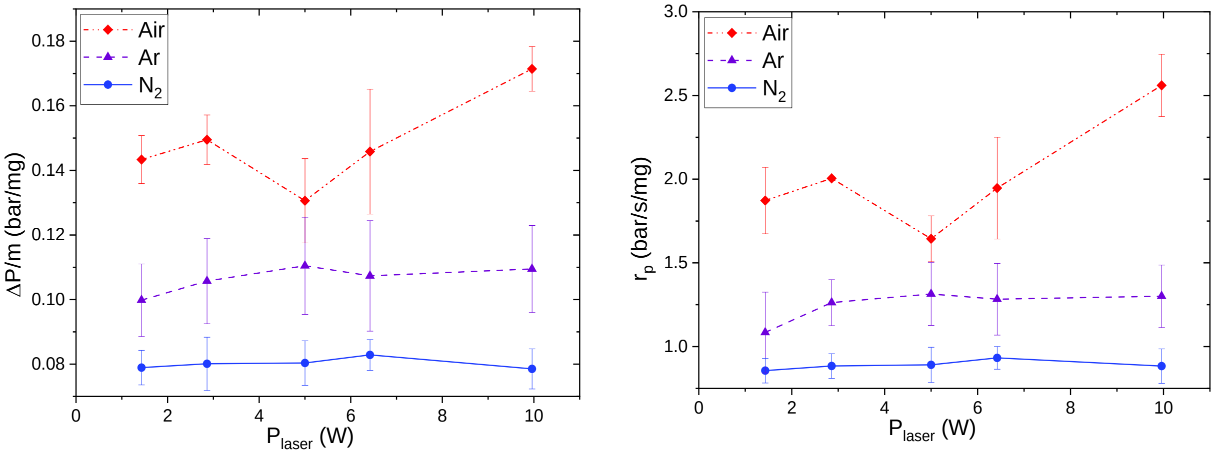


Figure 4: Ignition delays as functions of initial pressure and atmosphere, P_{laser} = 2.86 W, t_{pulse} = 500 ms

Influence of laser power and atmosphere

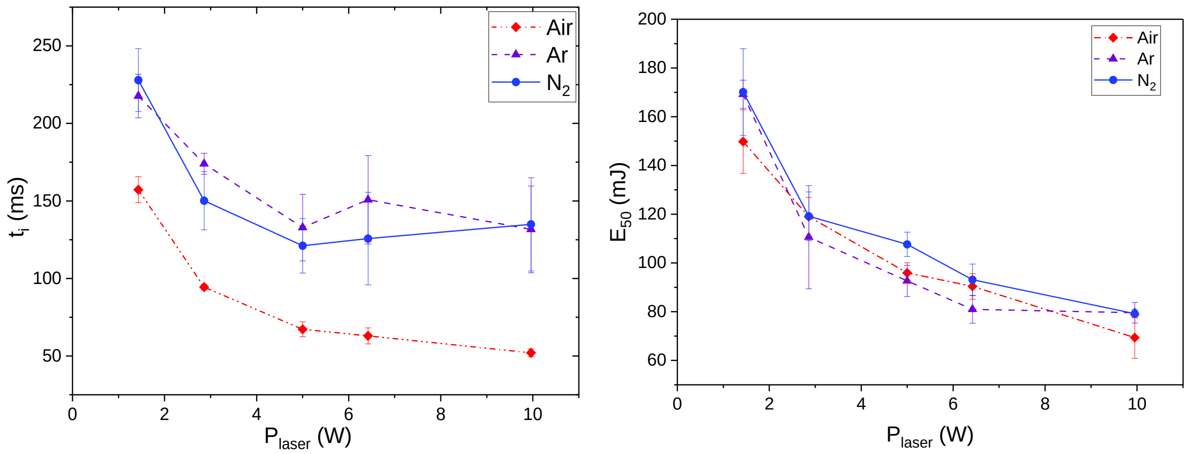
Figure 5 presents overpressures (a) and propagation rates (b) as functions of laser power at P_0 = 50 bar. These combustion properties are almost independent of the laser power for argon and nitrogen atmospheres. Under air, this assertion is particularly right between 1.43 W and 6.42 W. But at 9.95 W, however, overpressure increases by 17 % and propagation rate by 29 % in comparison with the previous values. This fact could be the sign of a threshold from which decomposition and reactions would be emphasized. Here, the influence of atmosphere is obvious with best properties for air followed by argon.



(a) *(b)*

Figure 5: Overpressures and propagation rates as functions of laser power and atmosphere, P_0 = 50 bar, t_{pulse} = 500 ms

Concerning ignition delays, presented Figure 6 (a), there is a clear effect of laser power until P_{laser} = 5.00 W, then, the t_i remain quasi-constant. The lowest ignition delays are obtained under air whereas results under nitrogen are better than argon. Figure 6 (b) finally presents E_{50} as functions of laser power for the three atmospheres at P_0 = 50 bar. Ignition energies under air, argon and nitrogen are not noticeably different and thus, E_{50} appear here to be independent of atmosphere. E_{50} are decreasing with an increase in power for the three gases.



(a) *(b)*

Figure 6: Ignition delays and probabilities as functions of laser power and atmosphere, P_0 = 50 bar

4 Conclusion

Ignition and combustion properties of an insensitive RDX-based gun propellant were experimentally investigated. Results especially focused on the influence of atmosphere on ignition delays, energies giving 50 % of probability of ignition, overpressures and propagation rates. Initial pressure and laser power were also studied as parameters. Combustion under synthetic air turned out to provide the best results, overcoming the negative oxygen balance of RDX, compared with argon and nitrogen. This experimental study is part of a wider project which aims to model ignition and combustion processes of low-vulnerability gun propellants using detailed kinetics chemistry for the gas phase. Thermal analysis experiments will also be conducted to investigate global kinetics of thermal decomposition for the condensed phase. The results presented here will thus enable the comparison with overpressures or burning rates provided by the model.

Acknowledgments

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References

[1] Melius CF. (1990). Thermochemical modeling, II: application to ignition and combustion of energetic materials. Chem Phys Energetic Mater: 51.

[2] Yetter RA, Dryer FL, Allen MT, Gatto JL. (1995). Development of gas-phase reaction mechanism for nitramine combustion. J. Propuls. Power. 11: 683.

[3] Prasad K, Yetter RA, Smooke MD. (1997). An eigenvalue method for computing the burning rates of RDX propellants. Combust Sci Technol. 124: 35.

[4] Liau YC, Kim ES, Yang V. (2001). A comprehensive analysis of laser-induced ignition of RDX monopropellant. Combust Flame. 126: 1680.

[5] Beckstead WB, Puduppakkam K, Thakre P, Yang V. (2007). Modeling of combustion and ignition of solid-propellant ingredients. Prog. Energy Combust Sci. 33: 497.

[6] Patidar L, Thynell ST. (2017). Quantum mechanics investigation of initial reaction pathways and early ring-opening reactions in thermal decomposition of liquid-phase RDX. Combust Flame. 178: 7.

[7] Khichar M. (2021). Thermal decomposition and combustion modeling of RDX monopropellant and RDX-TAGzT pseudo-propellant. The Pennsylvania State University. PhD dissertation.

[8] Gillard P, Courty L, De Persis S, Lagrange JF, Boulnois C, Gökalp I. (2018). Combustion properties of a low-vulnerability propellant: an experimental and theoretical study using laser ignition. J. Energetic Mater. 36: 362.

[9] Fang X, Stone M, Stennett C. (2020). Pulsed laser irradiation of a nanoparticles sensitised RDX crystal. Combust Flame. 214: 387.

[10] Aduev BP, Nurmukhametov DR, Liskov IY, Tupitsyn AV, Belokurov GM. (2020). Laser pulse initiation of RDX-Al and PETN-Al composites explosion. Combust Flame. 216: 468.

[11] Langlie HJ. (1962). A Reliability Test Method For One Shot Items. Ford Motor Company Aeronutronic. USA. Publication n° U1792.

[12] Hiyoshi RI, Brill TB. (2002). Thermal Decomposition of Energetic Materials 83. Comparison of the Pyrolysis of Energetic Materials in Air versus Argon. Propellants Explos. Pyrotech. 27: 23.