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Multilinear analysis of influential parameters on the combustion velocity of ternary pyrotechnic compositions

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Abstract

Combustion velocity is an important parameter for the formulation of pyrotechnic compositions. It is affected by several parameters that are studied in this work. Pyrotechnic compositions studied in the present work are ternary compositions composed of an oxidiser, a metallic reducer and a binder. This binder is also an energetic molecule. These three components are respectively strontium peroxide (SrO₂), magnesium (Mg), and 2,4-Dinitroanisole (DNAN). These model ternary pyrotechnic compositions were designed and produced at Nexter Munitions facility and characterized in PRISME Laboratory in Bourges. Thermal degradation and experimental burning velocities were already published.

The influence of specific parameters on the evolution of combustion velocity of ternary pyrotechnic composition are investigated using multilinear analysis, with the help of dimensionless numbers. These parameters are the amount of each constituent (oxidiser, reducer, binder), porosity, magnesium granulometry, combustion enthalpy and thermal conductivity estimated by analytical laws.

Using the studied parameters, three dimensionless numbers were implemented. Their influence on the combustion velocity was assessed and confirmed. Multilinear regressions were performed using these parameters, and the dimensionless numbers, to explain their joint influence on the combustion velocity. Results showed different joint influences before or after the combustion velocity maximum as a function of equivalence ratios.

Keywords: Pyrotechnic compositions – Combustion velocity – Multilinear regression – Dimensionless numbers – Strontium peroxide – Magnesium – DNAN

Introduction

Pyrotechnic compositions are used both in military and civilian fields [1-5]. A critical characteristic of these compositions is the combustion velocity. Therefore, understanding of the physicochemical phenomena that take place during the ignition and the propagation of the combustion are of great importance [6,7,8]. This work studies the influence of specific parameters on the evolution of the combustion velocity through multilinear analysis. Studied pyrotechnic compositions were designed and made at Nexter company.

Pyrotechnic compositions studied are composed of an oxidiser, a metallic reducer, and an energetic molecule used both for energy supply and mechanical binding, respectively strontium peroxide (SrO₂), magnesium (Mg), and 2,4-Dinitroanisole (DNAN). Raw materials and pyrotechnic compositions were thermally characterized using several techniques, e.g. DSC and calorimetric bomb [9]. The combustion velocities in small trough were also measured [10]. The studied parameters are the true and bulk densities, thermal conductivity, combustion enthalpy, magnesium granulometry and heat capacity of the pyrotechnic compositions. These parameters were used to obtain 2 dimensionless numbers, which are representative of the evolution of the combustion velocity. Multilinear regressions were then performed using these parameters and the dimensionless numbers to explain their joint influence on the combustion velocity.

Materials and methods

Pyrotechnic materials

The pyrotechnic compositions used in the present work are made of a SrO₂ rate between 47.5 wt% and 81 wt%, a Mg rate between 9 wt% and 47.5 wt% and a DNAN rate between 5 wt% and 10 wt%. Moreover, three granulometries of magnesium powder were used and named A, B and C for the following D₅₀:

- 140 µm < A < 200 µm

- 200 µm < B < 250 µm

- 250 µm < C < 300 µm

20 compositions were formulated for each magnesium granulometric class, leading to a total of 60 pyrotechnic compositions. They were analysed by Differential Scanning Calorimetry (DSC), Thermogravimetric Analysis (TGA) and calorimetric bomb in previous publications [9-11]. This work uses these previous results and the measured burning velocities gathered in Tables 1 and 2, and illustrated in Figures 1 to 3.

Table 1: Pyrotechnic compositions formulations.

Composition	wt.% Mg	wt.% SrO ₂	wt.% DNAN	Ox/Red Ratio (%)
X1	47.5	47.5	5	50
X2	34.2	60.8	5	64
X3	32.3	62.7	5	66
X4	28.5	66.5	5	70
X5	23.75	71.25	5	75
X6	19	76	5	80
X7	14.25	80.75	5	85
X8	13.49	81.51	5	85.8
X9	11.4	83.6	5	88
X10	9.5	85.5	5	90
X11	45	45	10	50
X12	32.4	57.6	10	64
X13	30.6	59.4	10	66
X14	27	63	10	70
X15	22.5	67.5	10	75
X16	18	72	10	80
X17	13.5	76.5	10	85
X18	10.8	79.2	10	88.78
X19	10.1	79.9	10	88
X20	9	81	10	90

Table 2: Pyrotechnic compositions burning velocities.

Composition	A		B		C	
	V (cm.s ⁻¹)	Porosity ϵ	V (cm.s ⁻¹)	Porosity ϵ	V (cm.s ⁻¹)	Porosity ϵ
X1	12.5	0.59	9.69	0.58	6.93	0.58
X2	13.6	0.58	10.71	0.65	10.83	0.62
X3	15.29	0.64	11.84	0.63	9.47	0.65
X4	16.82	0.64	11.71	0.61	8.59	0.61
X5	12.69	0.64	12.67	0.66	7.25	0.63
X6	14.5	0.65	7.28	0.66	6.06	0.62
X7	12.54	0.68	5.49	0.68	5.42	0.66
X8	10.89	0.64	6.18	0.67	4.49	0.67
X9	8.25	0.67	4.99	0.71	3.92	0.64
X10	6.94	0.67	4.78	0.70	3.12	0.71
X11	6.59	0.57	4.73	0.58	4.74	0.57
X12	6.98	0.63	6.65	0.59	5.5	0.63
X13	9.55	0.64	6.83	0.62	6.07	0.63
X14	10.56	0.65	4.82	0.63	5.86	0.62
X15	9.97	0.64	5.33	0.64	5.24	0.63
X16	9.9	0.67	4.72	0.63	4.5	0.63
X17	5.51	0.66	3.73	0.65	4.39	0.65
X18	7.84	0.67	3.68	0.66	3.98	0.67
X19	6.27	0.66	4.04	0.66	4.11	0.67
X20	7.13	0.67	3.63	0.66	4.27	0.67

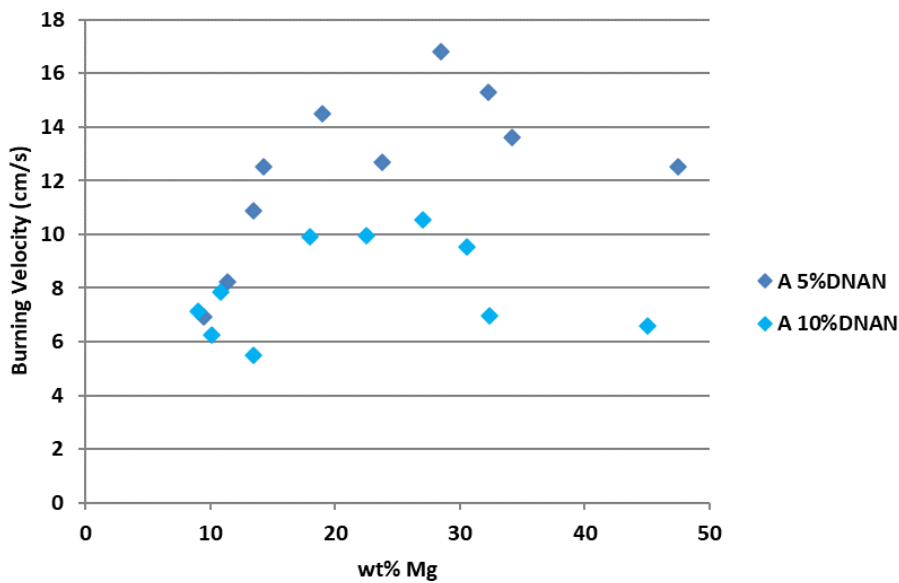


Figure 1: Burning velocities measured for pyrotechnic compositions A.

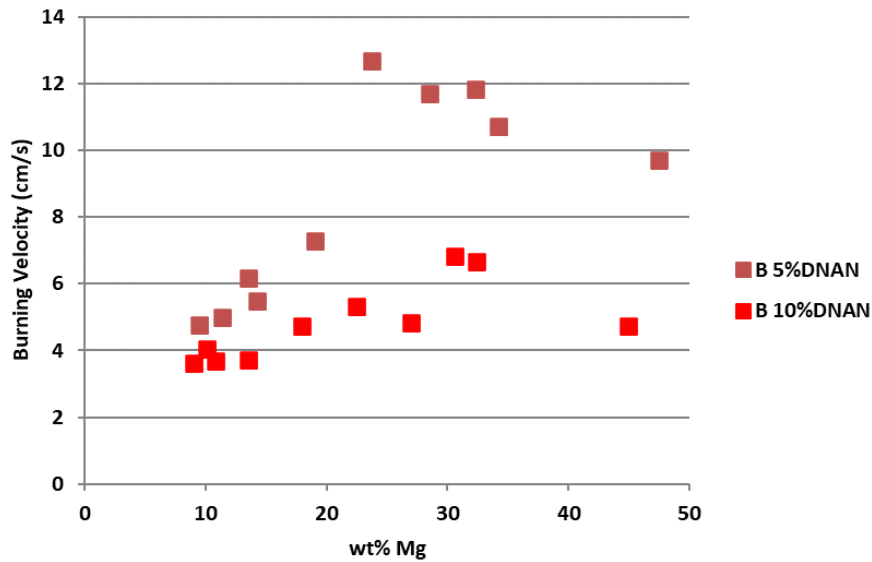


Figure 2: Burning velocities measured for pyrotechnic compositions B.

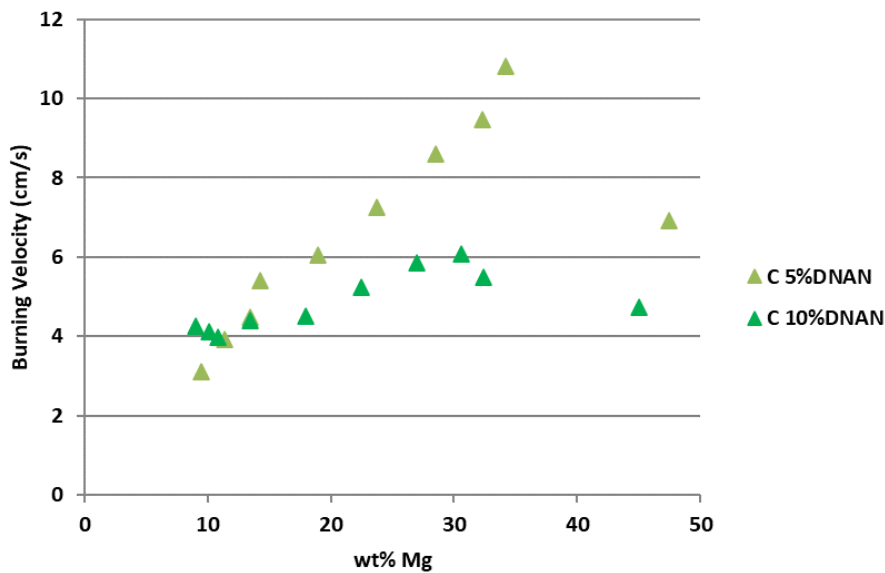


Figure 3: Burning velocities measured for pyrotechnic compositions C.

Dimensionless numbers

Six parameters having an influence on the burning velocity [5] have been selected. They are gathered in the Table 3 with their unit and physical dimension. Thanks to the Vaschy-Buckingham theorem (or Pi theorem), two dimensionless numbers were found and said to have good representation of the studied problem.

Table 3: Influential parameters studied.

Variable	Symbol	Unit	Physical dimension
Mg diameter	D_{50}	[m]	L
True density	ρ_{th}	[$kg \cdot m^{-3}$]	M, L^{-3}
Bulk density	ρ_{app}	[$kg \cdot m^{-3}$]	M, L^{-3}
Heat capacity	C_p	[$J \cdot kg^{-1} \cdot K^{-1}$]	$L^{-2}, T^{-2}, \Theta^{-1}$
Thermal conductivity	λ	[$W \cdot m^{-1} \cdot K^{-1}$]	$M, L, T^{-3}, \Theta^{-1}$
Reaction enthalpy	Q	[$J \cdot kg^{-1}$]	L^2, T^{-2}

The dimensionless numbers N_i obtained are:

- $\frac{\rho_{app}}{\rho_{th}}$: The ratio between the bulk density ρ_{app} , and the true density ρ_{th} , which is very close to the expression of the porosity ε , being $\varepsilon = 1 - \frac{\rho_{app}}{\rho_{th}}$.
- $\frac{\lambda}{\sqrt{Q} \cdot c_p \cdot D_{50} \cdot \rho_{app}}$: this dimensionless number N describes the ratio between the diffusive contribution and the enthalpic contribution. Using the expression of the thermal diffusivity α , it can be reduced as follow $\mathcal{N} = \frac{\alpha}{\sqrt{Q} \cdot D_{50}}$

The chemistry and formulation of the pyrotechnic compositions should already be considered through the determination of the enthalpy of the reaction Q and the thermal diffusivity α . Nevertheless, the authors think judicious to use the equivalence ratio Φ .

Φ_1 and Φ_2 are defined as the equivalence ratio of the reaction between the magnesium and the strontium peroxyde and the equivalence ratio between the DNAN and the strontium peroxyde respectively.

$$\Phi_1 = \Phi_{Mg/SrO_2} = \frac{m_{Mg,i}/m_{SrO_2,i}}{m_{Mg,st.}/m_{SrO_2,st.}}$$

$$\Phi_2 = \Phi_{DNAN/SrO_2} = \frac{m_{DNAN,i}/m_{SrO_2,i}}{m_{DNAN,st.}/m_{SrO_2,st.}}$$

The authors then define the ratio between Φ_1 and Φ_2 that allows to appreciate the influence of one reaction on the other:

$$\frac{\Phi_1}{\Phi_2} = \frac{Mg_i}{DNAN_i} \times \frac{m_{DNAN,st.}/m_{SrO_2,st.}}{m_{Mg,st.}/m_{SrO_2,st.}}$$

Results

The dimensionless number N is compared to the $\frac{\Phi_1}{\Phi_2}$ ratio in the following figures.

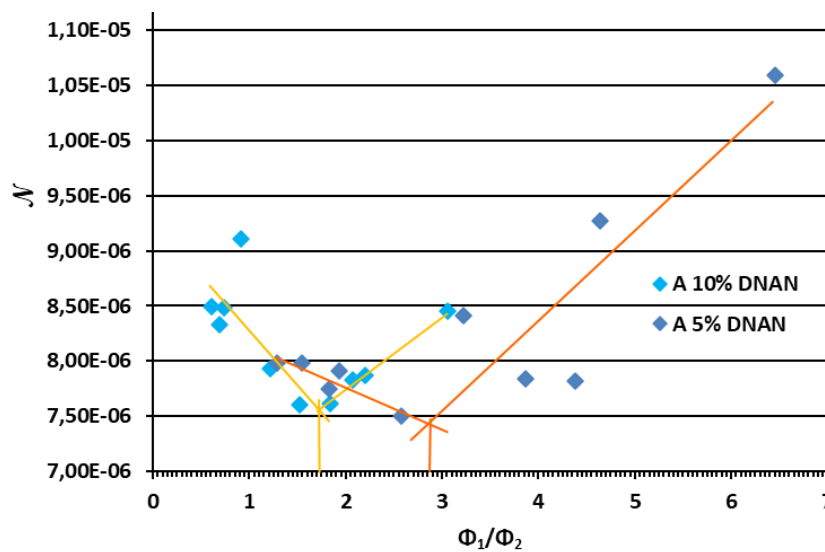


Figure 4: Extraction of supposed maximum burning composition formulation (A).

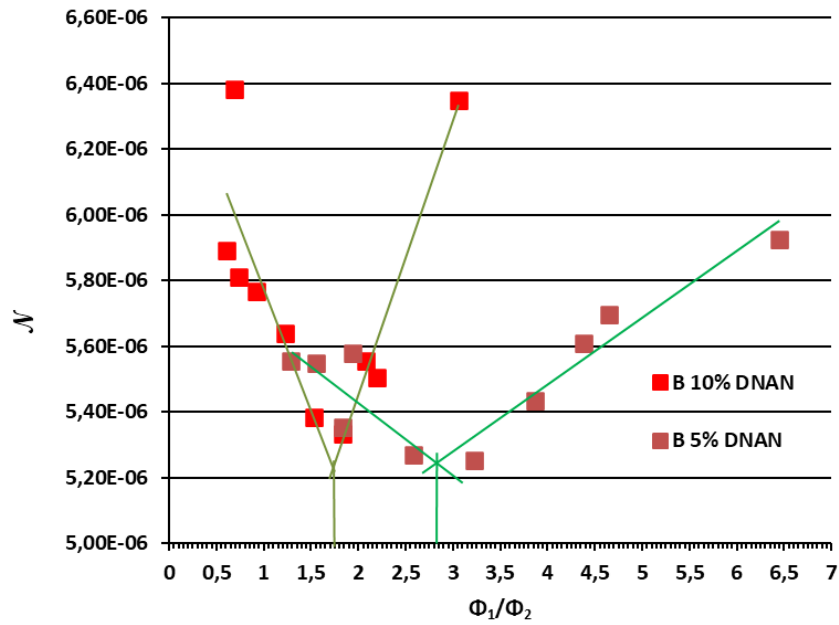


Figure 5: Extraction of supposed maximum burning composition formulation (B).

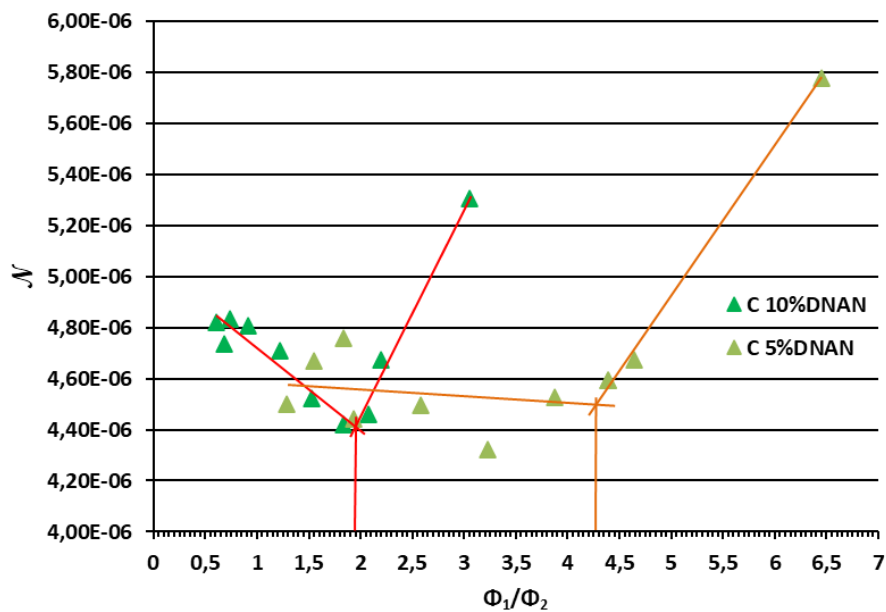


Figure 6: Extraction of supposed maximum burning composition formulation (C).

The obtained curves present a “V” shape with a local minimum corresponding to a certain amount of magnesium. The corresponding weight percentage of magnesium is extracted in the table below.

Table 4: Magnesium content for supposed maximum burning velocity composition.

Composition	Φ_1/Φ_2	wt% Mg
A 5 wt% DNAN	2.86	21.04
A 10 wt% DNAN	1.72	25.28
B 5 wt% DNAN	2.84	20.90
B 10 wt% DNAN	1.75	25.72
C 5 wt% DNAN	4.27	31.47
C 10 wt% DNAN	1.95	28.74

When compared with the measured burning velocities [9,11], it appears that the characteristic point obtained through the figures F4-6 would be the magnesium ratio of a pyrotechnic formulation that would produce the maximum of burning velocity, as it is close to the one observed experimentally. The obtained magnesium ratios are far from the stoichiometric ratios of magnesium which would be 13.49 and 10.10 wt% for compositions containing 5 and 10 wt% of DNAN respectively. It goes accordingly with scientific literature stating that the maximum of burning velocity is observed for compositions containing more reducer than the stoichiometric one. [12-13]

Multilinear regressions on dimensionless numbers

In order to study and quantify the influence of different parameters on the burning velocity of pyrotechnic compositions, we make the assumption that these parameters are linked by a power law. Each parameter has its own power coefficient such as the Vieille Law which links the burning velocity to the pressure:

$$V = a \cdot P^n$$

A generic law would be:

$$V = \prod_{i=1}^n C_i \cdot x_i^{a_i}$$

With x_i being the i^{th} influent considered parameter and a_i the power coefficient associated with, and C_i its associated constant.

These power coefficients a_i can be easily obtained using natural logarithm but doing so implies dimensionless values. x_i^* is the nondimensional x_i and is defined as:

$$x_i^* = \frac{\ln(x_i) - \ln(x_{i,\min})}{\ln(x_{i,\max}) - \ln(x_{i,\min})}$$

The selected parameters are studied through the dimensionless numbers N_i presented above and the linearisation then goes as follow:

$$V^* = b^* + \sum_{i=1}^4 a_i \cdot \ln(N_i)$$

With b being a constant gathering all remaining parameters that couldn't be considered.

The coefficients' matrix and associated curves are presented below.

Table 5: coefficients' matrix for dimensionless numbers alone.

R^2		b	N	$\rho_{\text{app}}/\rho_{\text{th}}$	Φ_1	Φ_2
0.712	N_i	6.02230	0.53317	-0.59952	0.29747	-0.32887
	N_1				0	0
	N_2	$\rho_{\text{app}}/\rho_{\text{th}}$			0	0
	N_3	Φ_1				0
	N_4	Φ_2				

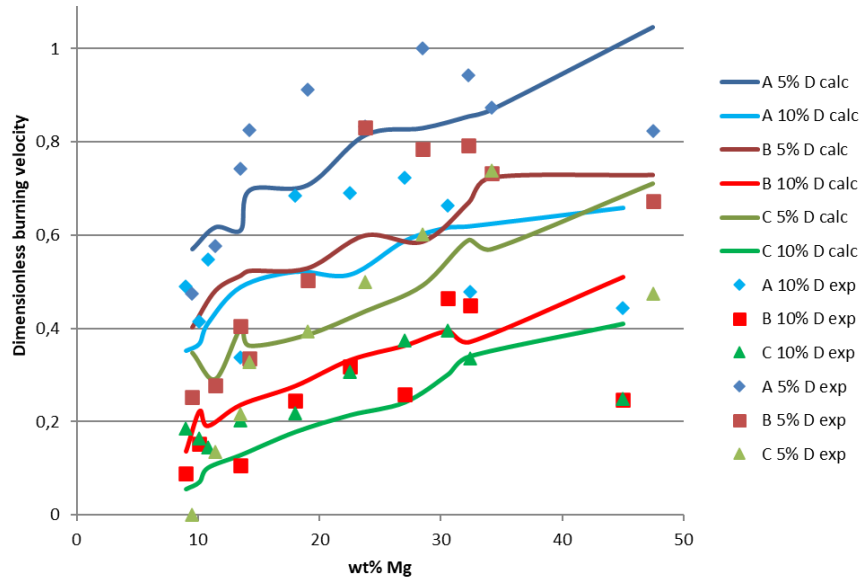


Figure 7: Comparison between measured dimensionless combustion velocities and calculated through multilinear regression (dimensionless numbers alone).

The values in the coefficients' matrix show that the equivalence ratio Φ_2 (which considers the reaction between SrO_2 and DNAN) has a negative influence on the increase of the burning velocity. That is to say, increasing the prevalence of this reaction will imply a diminution in burning velocity values, which goes accordingly with saying that increasing the quantity of binder has a negative influence on the burning velocity. In opposition, equivalence ratio Φ_1 has a positive influence on the increase of burning velocity. Densification ρ_{app}/ρ_{th} , which is the opposite of the porosity ε , has a negative influence too with a value of -0.59952 .

The coefficient of the number \mathcal{N} is positive with a value of 0.53317. This leads to two conclusions, the first is that being in the denominator, the particle size D_{50} , increases the value of \mathcal{N} while diminishing, thus has a positive influence on the increase in the burning velocity, which goes accordingly with the literature [5,13]. The second conclusion could be that the diffusive input α could have a greater, positive, influence than the enthalpic input \sqrt{Q} .

Nevertheless, the correlation coefficient R^2 is quite weak with a value of 0.712.

A complexification of the model is to consider the interactions between the dimensionless parameters. This new crossterm parameter is $N_{i,j}^*$:

$$N_{i,j}^* = N_i^* \times N_j^*$$

With $a_{i,j}$ its associated coefficient. It is important to note that the squares parameters e.g. $N_{i,i}^*$ and associated coefficients are not considered.

The following analysis presents the integration of the crossterms parameters with the matrix and curves.

Table 6: Coefficients' matrix for dimensionless numbers and crossterms.

	b	N	ρ_{app}/ρ_{th}	Φ_1	Φ_2	
R^2 0.871	Ni	-7.58790	-0.64441	-13.87073	-1.14322	-0.07072
N ₁	N		-1.14040	-0.04116	-0.02095	
N ₂	ρ_{app}/ρ_{th}			-0.83127	0.18566	
N ₃	Φ_1				-0.18079	
N ₄	Φ_2					

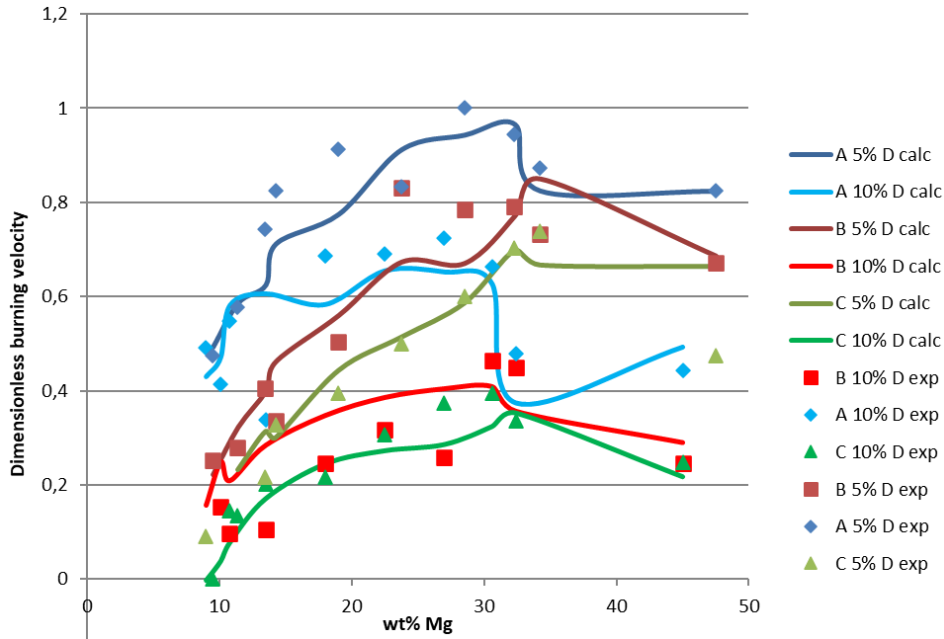


Figure 8: Comparison between measured dimensionless combustion velocities and calculated through multilinear regression (dimensionless numbers and crossterms).

The introduction of the crossterms parameters allows an increase in the model precision with a correlation parameter R^2 of 0.871.

The number $\frac{\rho_{app}}{\rho_{th}}$, still negative, becomes the driving parameter with a value of -13.87. This denotes the essential part of the porosity ($\varepsilon = 1 - \frac{\rho_{app}}{\rho_{th}}$), and the densification, on the burning velocity. Here it implies that porosity has a positive influence on the burning velocity. It means that a negative coefficient for the densification implies that, in the case of pyrotechnic composition, a powder bed too dense is not suitable for a greater burning velocity.

This assertion goes accordingly with another significant coefficient, the one linking $\frac{\rho_{app}}{\rho_{th}}$ and Φ_1 (reaction SrO_2/Mg) with a value of -0.831. Indeed, with too low porosity, the decomposition gases of the oxidiser cannot penetrate the bed further.

On the contrary, the sole term coefficient of Φ_2 seems negligible, with a value of -0.07. Its importance might already be taken into account through its reaction enthalpy in the term of \mathcal{N} . Its value is still negative, as increasing the binder content tends to diminish the burning velocity.

Though, Φ_2 has a positive influence with an increase of the term $\frac{\rho_{app}}{\rho_{th}}$, in opposition with $\frac{\rho_{app}}{\rho_{th}}$ and Φ_1 , meaning the reaction can increase the burning velocity in case of low porosity.

The negativity of the coefficient linked to Φ_1 is odd considering it being the main chemical reaction to take place. It could be explained by the fact that the studied reaction always has a magnesium content under 50 wt%, thus implying that a low equivalence ratio is positive to increase the burning velocity of the pyrotechnic composition.

The use of dimensionless parameters seems to allow a correct understanding of the known influences of the selected parameters on the burning velocity, but as showed in figs 4, 5, 6 regarding $\mathcal{N} = f\left(\frac{\Phi_1}{\Phi_2}\right)$, with "V" shape curves, different behaviours could be explained before and after the maximum value of burning velocity. The previous model will now be applied to the two parts of the curves.

Multilinear regressions before and after maximum burning velocity

The following figures present the multilinear regressions using the same method developed earlier and applied to the left and right branch of the respective « V » shaped curve. Tables 7 and 8 present the respective crossterms matrixes.

Table 7: coefficients' matrix for crossterms parameters of the left branches.

R^2		b	N	ρ_{app}/ρ_{th}	Φ_1	Φ_2
0.895	Ni	-31.78501	-2.47018	-33.88507	7.86460	-10.96939
	N ₁			-2.64045	0.42414	-0.61517
	N ₂				2.07954	-2.91100
	N ₃					-0.16277
	N ₄					

Table 8: coefficients' matrix for crossterms parameters of the right branches

R^2		b	N	ρ_{app}/ρ_{th}	Φ_1	Φ_2
0.889	Ni	9.81577	0.72325	1.28399	-2.92920	2.64993
	N ₁			0.06257	-0.17959	0.10675
	N ₂				-0.73347	1.40425
	N ₃					-0.32585
	N ₄					

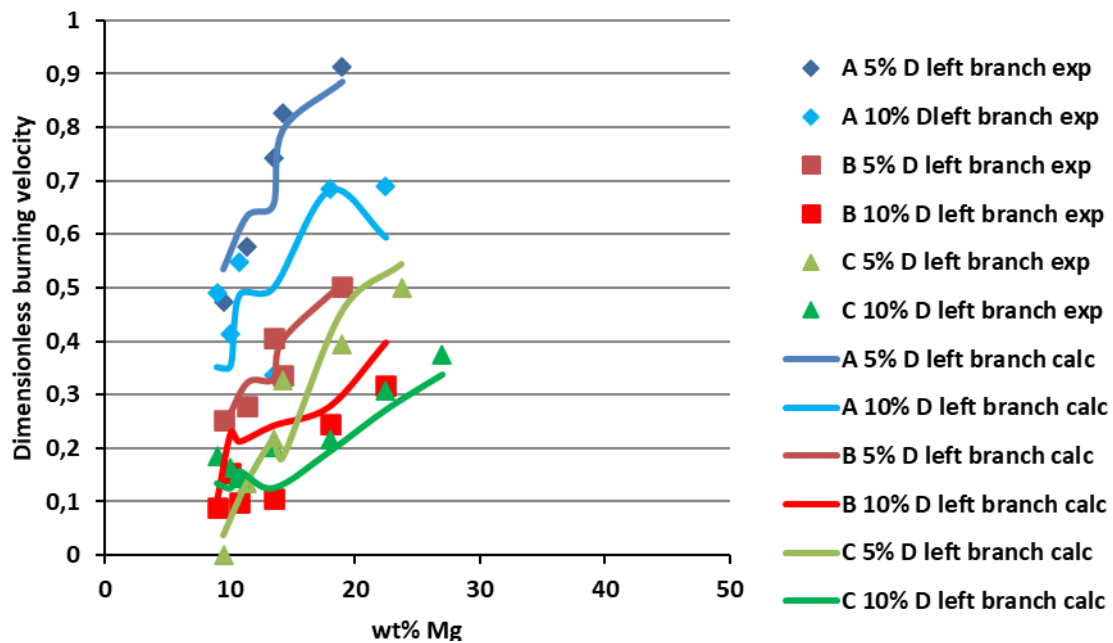


Figure 9: Comparison between measured dimensionless combustion velocities and calculated through multilinear regression (dimensionless numbers and crossterms) for left branches.

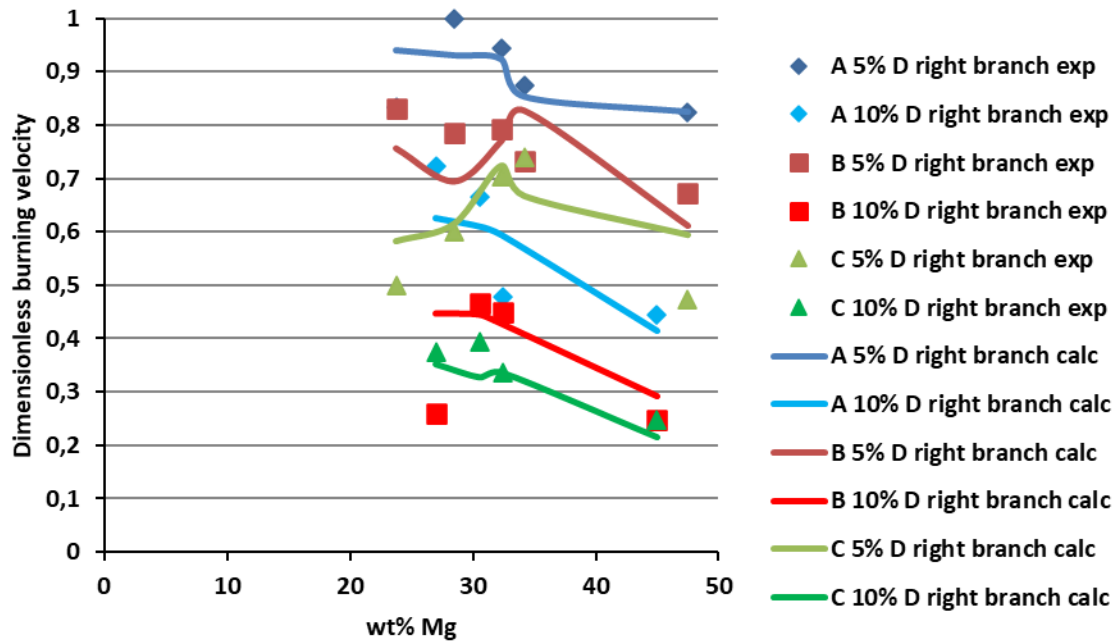


Figure 10 : Comparison between measured dimensionless combustion velocities and calculated through multilinear regression (dimensionless numbers and crossterms) for right branches.

The left branch (after the maximum burning velocity) and right branch (before the maximum burning velocity) have good enough correlation coefficients R^2 , 0.895 and 0.889 respectively, to assess that the model is correct

In the case of left branches, in fig 9 and Table 7, the coefficient of \mathcal{N} is negative with a value of $-2,47$. This means that the increase in \mathcal{N} has a negative influence on the increase in the burning velocity. \mathcal{N} value can increase through several ways. One is an increase in the thermal diffusivity α , another is a diminution in the reaction enthalpy (\sqrt{Q}) or finally a diminution in the particle size D_{50} . But the last two ways have a positive influence on the increase in the burning velocity. Then, for the same kind of pyrotechnic composition, an increase in the thermal diffusivity drives a decrease in the burning velocity. This goes accordingly with the other parameters. Indeed, the coefficient of Φ_1 is positive meaning that in the case of the left branches, an increase in Mg content allows greater burning velocities (Mg content is between 10 and 30 wt%). In fact, a greater thermal diffusion, in the case of SrO_2/Mg reaction driven compositions, is negative to the propagation of a combustion front. This is validated by the negative coefficient of the crossterm linking \mathcal{N} and ρ_{app}/ρ_{th} .

Indeed, the sole term coefficient of ρ_{app}/ρ_{th} is negative with a value of -33.89 , meaning that a densification of the bed is negative to an increase in burning velocity. The left branches compositions are driven by an increase in porosity.

The negative value of the coefficient of Φ_2 goes accordingly with a negative influence of the binder content, through the reaction SrO_2/DNAN , on the burning velocity.

In the case of right branches compositions, the coefficient associated with the equivalence ratio Φ_1 of the reaction SrO_2/Mg is negative. Indeed, maximum velocity has already been attained and an increase in magnesium content will only imply a diminution of the burning velocity by increasing the thermal diffusivity. It is validated by the crossterm linking Φ_1 and ρ_{app}/ρ_{th} being negative (-0.73). In contrast, the coefficient of Φ_2 is positive, meaning that the increase in DNAN allows a diminution in the thermal conductivity thus a decrease in thermal diffusivity. Finally, ρ_{app}/ρ_{th} having a small value contrary to the other parameters and not being the driving parameter as in the left branches compositions is odd. Moreover, considering it presents a coefficient with a positive value is unexpected, as porosity could help diminish the thermal diffusivity.

The positivity of the coefficient of \mathcal{N} can be attributed to the positive influence of the particles size.

Conclusion

The present article presents the joint influence of different parameters on the burning velocities of ternary pyrotechnic compositions containing SrO_2 as an oxydiser, Mg as a metallic reducer and DNAN as an energetic binder.

First, the selected parameters were used to obtain 2 dimensionless numbers to describe the problem through Vaschy-Buckingham theorem (Pi theorem). These two dimensionless numbers are ρ_{app}/ρ_{th} and \mathcal{N} ($\mathcal{N} = \frac{\alpha}{\sqrt{Q \cdot D_{50}}}$), and the equivalence ratios of the chemical reactions SrO_2/Mg and SrO_2/DNAN : Φ_1 et Φ_2 were added to the study. The study of these dimensionless numbers, especially $\mathcal{N} = f\left(\frac{\Phi_1}{\Phi_2}\right)$ "V" shaped curves allowed, thanks to a graphical method, the extraction of the formulation that could produce the maximum of burning velocity for that specific pyrotechnic composition.

Second, the assumption was made that like the Vieille Law $V = a \cdot P^n$, the burning velocity could be represented following a power law using the selected parameters. Following that goal, the dimensionless numbers were used to assess their influence on a nondimensionalised burning velocity V^* of the ternary pyrotechnic compositions. Then, a multilinear regression was used to extract and study the coefficients of these dimensionless numbers. The first low precision model ($R^2=0.712$) lead to a complexification of the model was made using crossterm coefficient to study the cross influence of the dimensionless numbers. The precision of the model was increased with $R^2=0.871$. It was demonstrated that porosity, through the densification term ρ_{app}/ρ_{th} , was the driving parameter of the ternary pyrotechnic compositions.

Finally, this method was applied to the left and right branches of the "V" shaped curves of $\mathcal{N} = f\left(\frac{\Phi_1}{\Phi_2}\right)$ to determine the different ways of influence of the selected parameters on the burning velocity on both sides of the maximum measured. It was effectively demonstrated, through the study of the dimensionless numbers, that the driving chemical reaction changed depending on which side of the maximum burning velocity we are located.

However, it is necessary to note that the parameters used are linked together by complex mixing laws, it is then possible that these interactions might have an influence on the value of the obtained coefficients.

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