



*Cent. Eur. J. Energ. Mater.* 2020, 17(2): 223-238; DOI 10.22211/cejem/122326

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*Research paper*

## Thermochemical Properties, Ballistic Parameters and Sensitivity of New RDX-based Propellants

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**Abstract:** New RDX-based propellants were obtained and investigated. The heat of combustion was determined in a calorimeter and the pressure history was measured in a manometric bomb. Ballistic parameters, such as maximum pressure, propellant force and covolume of gaseous products, as well as the dynamic vivacity, were determined based on the experimental results. The sensitivities to friction and impact were determined for the tested propellants. Calculations of the ballistic characteristics (the heat of combustion, propellant force, covolume and ratio of specific heats of the combustion products, and ballistic energy) were performed. The theoretical parameters were compared with the experimental data and quite good agreement was found between them. The largest differences occurred in the cases of propellant force and covolume. This disagreement is explained. An analysis of the test results and a comparison of the parameters of the RDX-based propellants and the JA2 propellant allowed the possibility of using the tested propellants in ammunition to be assessed.

**Keywords:** RDX-based propellants, ballistic parameters, thermochemical model

## 1 Introduction

Low sensitivity propellants usually consist of 60-85% nitramine filler (RDX, HMX) as an oxidant, 10-25% of a polymeric binder and one or more plasticizers [1]. These propellants are often used in low-vulnerability ammunition (LOVA). Examples of low sensitive propellants are XM39 and M43 [2, 3]. In both of these propellants, cyclo-1,3,5-trimethylene-2,4,6-trinitramine (RDX) is the main high energy component, with cellulose acetate butyrate (CAB) as the binder. The combustion characteristics and thermochemical properties of these two types of RDX-based propellants, XM39 and M43, were studied in [4]. The burning rate exponents and coefficients, surface temperatures, activation energies and exponents as deduced from the experimental results were compared. Systematic studies on the influence of the RDX particle size on the burning rate and ballistics of LOVA gun propellants were carried out in [5]. The propellant composition consisted of RDX 78%, cellulose acetate 12%, nitrocellulose (NC) (12.3% N) 4%, glycerin triacetate (Triacetin) 5.85% and ethyl centralite 0.2%. Fine RDX particles, with a size of 4.5, 6, 13 and 32  $\mu\text{m}$ , were used. The authors concluded that the particle size of fine RDX played a significant role in determining the ballistic parameters of the propellants. Moreover, fine RDX of about 4.5  $\mu\text{m}$  particle size can be used for LOVA propellant formulations. RDX's influence on the performance increase of triple base propellants was studied in [6]. The triple base composition included NC, nitroglycerine (NG) and picrite. From thermochemical calculations and closed vessel experiments, it can be concluded that partial replacement of picrite by RDX (from 5 to 20%) in the base propellant causes an increase in chemical energy as well as an improvement in ballistic performance. Although approximate compositions and methods for the production of low-sensitivity multi-base propellants are the focus of numerous publications in scientific journals, conference communications and patents, this does not mean that it is possible to perform a simple reproduction of already developed technologies.

In the present work, new RDX-based propellants of low sensitivity were obtained and tested. Some of these propellants were used in [7], to verify the thermochemical model and calculation conditions for the determination of the heat of combustion of propellants. In present paper, the pressure histories and heats of combustion were measured for all of the propellants. The sensitivities to friction and impact were also determined. The CHEETAH code with the BLAKE product library [8] was used to calculate the ballistic parameters of the propellants, including their heat of combustion. The composition of the new propellants was changed in such a way that the ballistic characteristics

of the final propellant, in particular the rate of pressure build-up in a manometric chamber and dynamic vivacity, were as close as possible to the characteristics of the JA2 propellant. The results of the work may be used to develop the technology of new low-sensitivity propellants.

## 2 Experimental

### 2.1 Preparation of the propellants

RDX of reduced sensitivity (RDX-RS), with particle size from the range 2.5-5 mm, was used for the preparation of the propellants. The other ingredients of the powders were: NC (12.6% N), CAB, Triacetin, Akardite II, soot and a burn-rate modifier. The compositions of the tested propellants are listed in Table 1. Grains of the propellants P1 to P3 were monolithic and P4 to P6 propellant grains had seven channels. For comparison, the tests and calculations were also made for the JA2 with the following composition:

- NC (13.2% N) – 58.21%,
- NG – 15.79%,
- diethyleneglycol dinitrate (DEGDN) – 25.18%,
- Akardite II – 0.74%,
- MgO – 0.05%,
- graphite – 0.03%.

The JA2 grains had seven channels.

**Table 1.** Composition of propellants selected for testing

Propellant symbol	Content of the component [wt.%]						
	RDX-RS	CAB	Triacetin	NC (12.6% N)	Akardite II	Soot	Modifier
P1	75.8	12.1	7.7	4.0	0.4	–	–
P2	76.0	–	7.6	16.0	0.4	–	–
P3	76.0	6.0	7.6	10.0	0.4	–	–
P4	76.0	6.0	7.6	10.0	0.4	–	–
P5	75.0	6.0	6.6	10.0	0.4	0.3	1.7
P6	75.3	6.1	6.4	10.0	0.4	0.3	1.5

The solid and liquid ingredients were mixed in a standard sigma blender with stainless steel blades for 4 h. The propellant grains were then formed with a standard laboratory extruder. Pictures of typical propellant grains are shown in Figure 1.



**Figure 1.** Pictures of P4 and P5 grains (grain diameter 7.6 mm)

## 2.2 Heat of combustion

The thermal effect accompanying the combustion reaction of the tested propellants was measured and recorded using a water calorimeter KL-10 produced by *PRECYZJA-BIT* from Bydgoszcz (Poland). A cylindrical steel calorimetric bomb with a capacity of approximately 350 cm<sup>3</sup> was placed in a steel vessel with a capacity of 4.4 dm<sup>3</sup> containing distilled water. The calorimetric system has been described in detail in [7].

The measurements of the heat of combustion of the propellants were carried out in an argon atmosphere. The air in the bomb was removed by filling with argon three times to a pressure of 0.5 MPa and finally the argon was left under this pressure. A sample of propellant with a weight of approximately 3 g in the form of 4 cylindrical grains was bonded with a spiral wire and a 6 cm long cotton cord and placed in a quartz crucible. The heat of combustion of the resistance wire and cotton cord was not taken into consideration due to its insignificantly low value compared to the heat of combustion of the propellant sample. Three tests were carried out for each type of propellant. The results of the combustion heat measurements are listed in Table 2.

**Table 2.** Measurement results of the heat of combustion of the tested propellants

Propellant symbol	Heat of combustion [J/g]	Average heat of combustion [J/g]
P1	3504	3520 ±50
	3492	
	3570	
P2	4547	4560 ±20
	4554	
	4585	
P3	4044	4060 ±40
	4098	
	4039	
P4	3941	3960 ±40
	4002	
	3946	
P5	3893	3900 ±30
	3869	
	3931	
P6	3786	3780 ±30
	3754	
	3798	
JA2	4655	4660 ±20
	4666	
	4646	

The heat of combustion of propellant P-2 is comparable to that of JA2. Propellant P1 has the smallest heat of combustion. Other propellants have comparable heat of combustion values, and smaller than that of JA2.

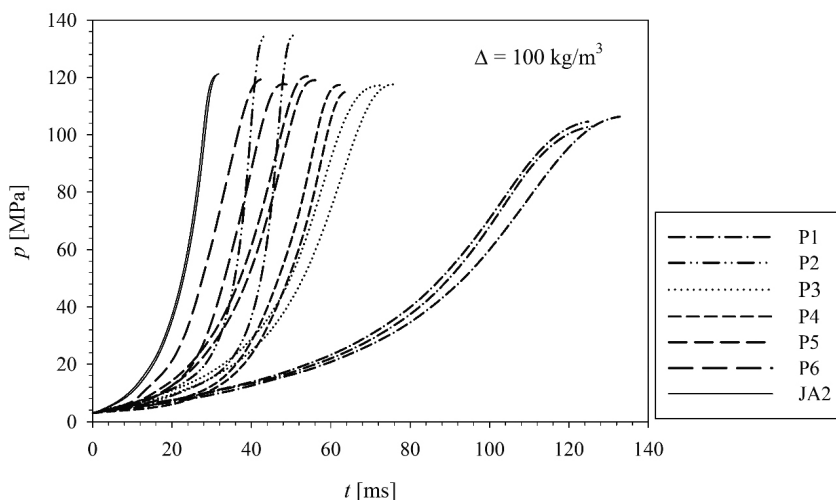
### 2.3 Closed vessel tests

The closed vessel tests of the propellants consisted of burning a specific powder mass ( $\omega$ ) in a constant volume ( $V$ ) (thick-walled cylindrical chamber) and measuring the pressure history ( $p(t)$ ) of the propellant gases. These histories are the basis for the determination of the ballistic characteristics of a propellant, such as the force ( $f$ ) and covolume ( $\alpha$ ) of the propellant gases. For this purpose the experimentally determined dependence of the maximum pressure ( $p_m$ ) of the gases on the loading density ( $\Delta = \omega/V$ ) was used. This dependence has the form of the Nobel-Abel formula (Equation 1).

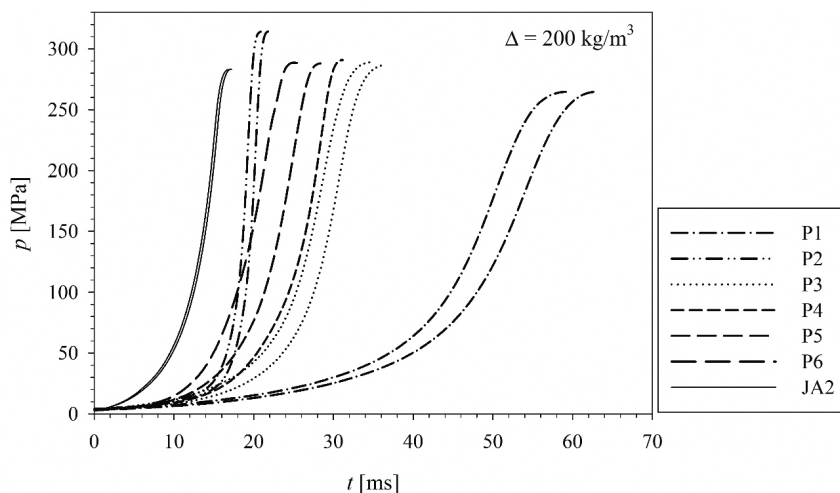
$$p_m = \frac{f \Delta}{1 - \alpha \Delta} \quad (1)$$

At least two values of  $p_m$ , determined for two different loading densities, enable  $f$  and  $\alpha$  to be calculated.

Closed vessel tests were performed for two values of the loading density:  $\Delta = 100$  and  $200 \text{ kg/m}^3$ , using a vessel with a capacity of  $200 \text{ cm}^3$ . The pressure was measured with a 5QP 60000M piezoelectric transducer manufactured by HPI-GmbH. The signal from the transducer was amplified by a TA-3/D amplifier and recorded on a Keithley DAS-50 12-bit analog-to-digital converter at a frequency of 1 MHz. Propellant charges were fired by an igniter having mass of 1.998 g (for  $\Delta = 100 \text{ kg/m}^3$ ) and 1.865 g (for  $\Delta = 200 \text{ kg/m}^3$ ), composed of black powder D-2 included in a small combustible foil bag. The ignition of the black powder was initiated by means of a thermal impulse emitted from the fuse activated by an electrical impulse. The pressure variation was sampled with time intervals of 25 ms. Pressure changes until the maximum value is reached are shown in Figures 2 and 3 for loading densities of 100 and  $200 \text{ kg/m}^3$ , respectively. The pressure build up curve determined for the P5 and P6 propellants are the closest to the curve of JA2.



**Figure 2.** Measured pressure histories for a loading density of  $100 \text{ kg/m}^3$

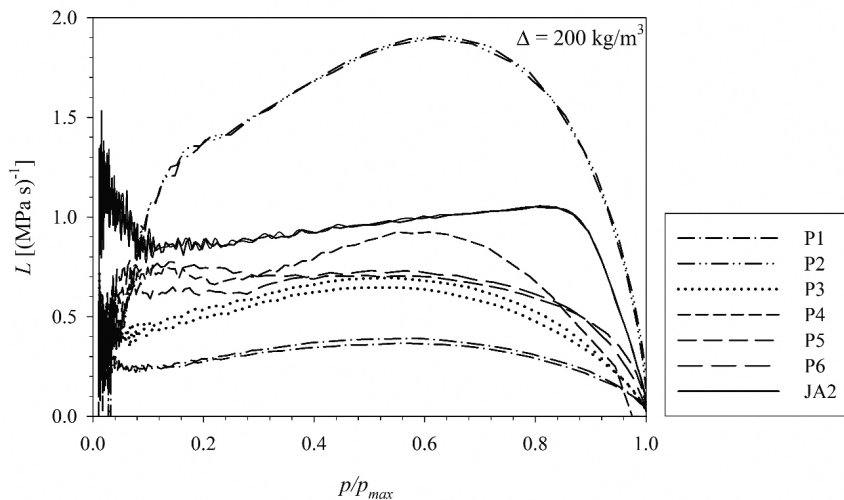


**Figure 3.** Measured pressure histories for a loading density of  $200 \text{ kg/m}^3$

The dependence of  $dp/dt$  on pressure for the tested propellants was obtained by using a spline method. This dependence was used to determine the dynamic vivacity [9] described by Equation 2:

$$L = \frac{dp}{dt} \frac{1}{p \cdot p_{max}} \quad (2)$$

where  $p_{max}$  is the maximum gas pressure of the smoothed pressure time curve. The dependence of the vivacity on relative pressure for a loading density of  $200 \text{ kg/m}^3$  is shown in Figure 4. The maximum vivacity for propellant P1 is the smallest of those tested, and the vivacity of P2 is the greatest. The vivacities of P2-P6 propellants are smaller than that determined for the JA2 propellant. However, they are comparable with the vivacity of other LOVA RDX-based propellants. The values of maximum pressures and corrected maximum pressures, obtained after taking heat losses into account, are listed in Table 3. All values (measured and corrected) of the maximum pressure were reduced by the ignition pressure of 3 MPa.



**Figure 4.** Dynamic vivacity for a loading density of  $200 \text{ kg/m}^3$

**Table 3.** Measurement results of the maximum pressure and corrected maximum pressure of the tested propellants

Propellant symbol	Loading density $\Delta$ [ $\text{kg/m}^3$ ]	Maximum pressure [MPa]	Corrected maximum pressure [MPa]
P1	100	103.5	116.3
		102.0	114.0
	200	261.8	271.9
		262.0	272.3
P2	100	131.8	135.7
		131.5	135.2
	200	311.3	314.1
		311.1	314.0
P3	100	114.2	123.6
		114.6	123.4
	200	283.6	292.1
		286.1	295.1
P4	100	112.2	124.2
		114.4	126.4
	200	287.9	298.2
P5	100	116.1	127.4
		117.4	129.0
	200	285.0	292.8



P6	100	114.7	125.5
		116.5	126.6
	200	281.3	287.1
JA2	100	118.3	127.4
		117.8	126.7
	200	280.2	287.6
		280.0	287.3

## 2.4 Sensitivity tests

The friction sensitivity of the tested propellants was determined on a Julius-Peters apparatus, according to the standard PN-EN 13631-3 [10]. A 0.5 mm thick disc of the tested sample was placed between a porcelain stamp and a porcelain plate in the Julius-Peters apparatus. The lowest loading at which a positive reaction (the occurrence of bang, crack, spark or flame smoke) was observed in at least one of six attempts was assumed as the sensitivity to friction. The impact sensitivity was determined using a BAM apparatus with a 5 kg hammer, according to the standard PN-EN 13631-4 [11]. A disc of 0.04 g weight of the tested propellant was used in each trial. The lowest height at which a positive reaction (occurrence of sound effects or a flame) was observed in at least one of six trials was taken as the sensitivity to impact. The results of the impact and friction sensitivity tests are shown in Table 4. The sensitivities for pure RDX and RDX phlegmatized by 6% of wax (RDX<sub>ph</sub>), in the form of powder, are included for comparison.

**Table 4.** Sensitivity to mechanical stimuli

Tested sample	Friction sensitivity [N]	Impact sensitivity [J]
RDX	165	3
RDX <sub>ph</sub>	250	7
P1	>360	16
P2	>360	12
P3	>360	10
P4	>360	8
P5	>360	10
P6	>360	10
JA2	230	4.5

The RDX-based propellants can be considered as insensitive to friction. The sensitivity to impact of the tested propellants was lower than that

determined for JA2 and RDXph. One of them, P1, had a sensitivity comparable to TNT (16 J). However, the impact sensitivity of the other composite propellants was satisfactory.

### 3 Calculation of the Ballistic Parameters

The CHEETAH code was used to calculate the ballistics parameters of the tested propellants. The thermodynamic model of combustion with the virial equation of state was implemented into this code [8]. The basic calculated ballistic parameters were:

- the heat of combustion in a constant volume,  $Q_v$ :

$$Q_v = \sum_i n_i (\Delta E_{(298.15)}^0)_i - \Delta E_s^0 \quad (3)$$

where  $n_i$  is the number of moles of the  $i$ -th component of the combustion products,  $\Delta E_{(298.15)}^0$  is the energy of formation of the  $i$ -th component, and  $\Delta E_s^0$  is the energy of formation of the propellant at constant volume,

- propellant force  $f$ :

$$f = n_g R T_v \quad (4)$$

where  $T_v$  is the isochoric combustion temperature,  $n_g$  – the number of moles of gaseous products,

- gas volume of combustion products  $\alpha$  occurring in the Nobel-Abel equation (covolume):

$$p(V - \alpha) = n_g R T_v \quad (5)$$

- ratio of the “frozen” specific heat capacities,  $\kappa$ :

$$\kappa = c_p / c_v \quad (6)$$

- ballistic energy  $E_b$ :

$$E_b = f / (\kappa - 1) \quad (7)$$

- pressure of the combustion products in a constant volume ( $p_v$ ).

The data used in the thermochemical calculations of the propellants are summarized in Table 5. The enthalpy of formation for CAB was determined from the experimental heat of combustion of this substance in an oxygen atmosphere.

**Table 5.** Data for thermochemical calculations of the propellants

Component	Chemical formula	Density [g/cm <sup>3</sup> ]	Molar mass [g/mol]	Molar volume [cm <sup>3</sup> /mol]	Enthalpy of formation [cal/mol]
CAB	C <sub>1,47</sub> H <sub>2,37</sub> O <sub>77</sub> [11]	1.25 [13]	3236.4 [12]	2589.2	-3 207 700
Akardite II	C <sub>14</sub> H <sub>14</sub> ON <sub>2</sub>	1.236	226.27	183.07	-25 500 [14]
Triacetin	C <sub>9</sub> H <sub>14</sub> O <sub>6</sub>	1.16 [13]	218.2 [13]	188.10	-318 070 [14]
DEGDN	C <sub>4</sub> H <sub>8</sub> O <sub>7</sub> N <sub>2</sub>	1.38	196.12	142.12	-99 400 [14]
NG	C <sub>3</sub> H <sub>5</sub> O <sub>9</sub> N <sub>3</sub>	1.60	227.1	142.29	-88 600 [14]
RDX	C <sub>3</sub> H <sub>6</sub> O <sub>6</sub> N <sub>6</sub>	1.81	222.1	122.99	14 710 [8]
NC (13.3% N)	C <sub>6</sub> H <sub>7,312</sub> O <sub>10,376</sub> N <sub>2,688</sub>	1.660	283.90	171.02 [8]	-163 460 [14]
NC (12.6% N)	C <sub>6</sub> H <sub>7,549</sub> O <sub>9,901</sub> N <sub>2,451</sub>	1.655	272.73	164.79 [8]	-169 171 [14]

**Table 6.** Results of calculations of the ballistic parameters

Propellant symbol	$Q_v$ [J/g]	$\alpha$ [cm <sup>3</sup> /g]	$f_p$ [J/g]	$c_p/c_v$	$E_b$ [J/g]	$T_v$ [K]	$p_v$ [MPa]	$n_g$ [mol/kg]
P1	3181	1.158	1083	1.267	4053	2712	282.0	48.05
P2	4103	1.070	1233	1.242	5094	3426	313.6	43.27
P3	3363	1.112	1167	1.254	4595	3076	300.3	45.65
P4	3663	1.112	1167	1.254	4595	3076	300.3	45.65
P5	3513	1.118	1137	1.258	4411	2976	297.6	45.96
P6	3853	1.117	1144	1.257	4449	2998	294.6	45.89
JAZ	4209	0.983	1161	1.224	5193	3486	289.0	40.04

The ballistic parameters of the propellants were calculated using the CHEETAH code for a loading density of 0.2 g/cm<sup>3</sup>. However, the heat of combustion was calculated as the difference between the standard enthalpy of formation of the products resulting from combustion under isochoric conditions and the enthalpy of formation of the propellant, *i.e.*:

$$Q_v = - \left[ \sum_i n_i (\Delta H_{(298.15)}^0)_i - \Delta H_s^0 - n_g R 298.15 \right] \quad (8)$$

where  $\Delta H_{(298.15)}^0$  is the enthalpy of formation of the *i*-th component, and  $\Delta H_s^0$  is the enthalpy of formation of the propellant at constant pressure. The work of the expansion of the combustion products ( $n_g R 298.15$ ) was added so that the calculated heat corresponded to Equation 3, in which the standard energies of formation are used. It was assumed that the water is in a gaseous state.

The results of the calculations of the ballistic parameters for the tested propellants are listed in Table 6. The results of these calculations from Table 6 indicate that some of the composite propellants have ballistic parameters comparable to those of the JA2 propellant.

## 4 Analysis of Measured and Calculation Results

Burning a propellant sample with a mass of approx. 3 g in the 0.35 dm<sup>3</sup>-volume bomb filled with argon at a pressure of 0.5 MPa is a complex process. To calculate the heat of combustion of a propellant corresponding to the calorimetric heat, in Ref. [7] the heat of combustion was calculated taking into account the argon filling the calorimetric bomb and the freezing of the composition of the products at different temperatures. Based on an analysis of the measured results and calculations, it was found that the best agreement between the calorimetric heat and the theoretical one was obtained when the composition of the combustion products is assumed to be frozen at 1300 K.

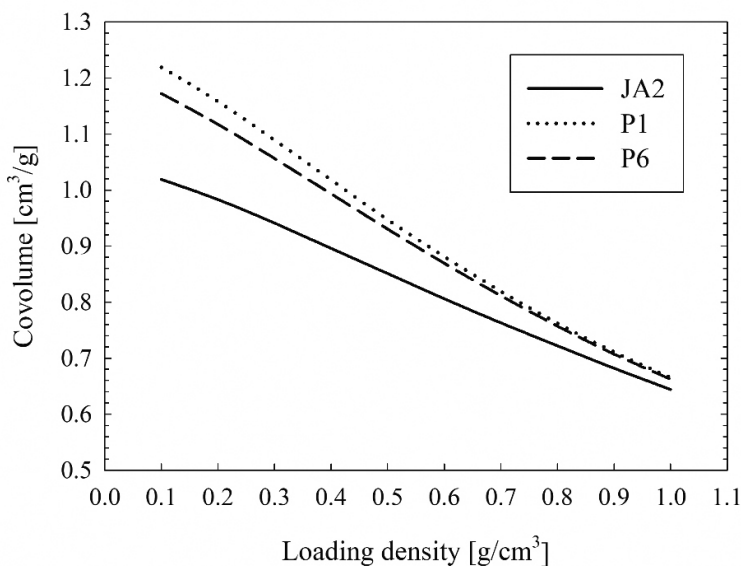
The CHEETAH code was used to determine the equilibrium state of reagents in the calorimeter volume. Then, the composition of the combustion products changing during cooling of the gaseous mixture in the calorimeter was frozen at 1300 K. The reaction heat ( $Q_z$ ) was obtained from Equation 8. Water was assumed to be in the liquid state in the calculations of  $Q_z$ . The comparison of the experimental ballistic parameters and those obtained from calculations are presented in Table 7.

**Table 7.** Comparison of the theoretical and experimental values of the ballistic parameters of the tested propellants

Propellant symbol	$Q_z$ [J/g]		$p_m$ [MPa] ( $\Delta = 100$ kg/m <sup>3</sup> )		$p_m$ [MPa] ( $\Delta = 200$ kg/m <sup>3</sup> )		$f_p$ [J/g]		$\alpha$ [cm <sup>3</sup> /g]	
	calc.	exp.	calc.	exp.	calc.	exp.	calc.	exp.	calc.	exp.
P1	3541	3520	123.6	115.2	282.0	272.1	1083	983	1.158	1.389
P2	4548	4560	138.4	135.5	313.6	314.1	1233	1191	1.070	1.208
P3	4040	4060	132.2	123.5	300.3	293.6	1167	1066	1.112	1.370
P4	4040	3960	132.2	125.3	300.3	298.2	1167	1080	1.112	1.377
P5	3880	3900	128.9	128.2	293.0	292.8	1137	1141	1.118	1.116
P6	3891	3780	129.6	126.1	294.6	287.1	1144	1079	1.117	1.241
JA2	4694	4660	128.5	127.1	289.0	287.5	1161	1138	0.983	1.040

The results from Table 7 indicate that there is quite good agreement between the experimental and calculated parameters. This means that the thermochemical model used in the CHEETAH code can be successfully used to optimize the composition of propellants from the point of view of the desired ballistic parameters.

Good compliance of the theoretical and experimental parameters was obtained for the heat of combustion  $Q_z$  and maximum pressure for a loading density of  $0.2 \text{ g/cm}^3$ . The largest differences occurred in the case of propellant force  $f$  and covolume  $\alpha$ . Large differences between the theoretical and experimental values of the propellant force are the result of the methodology used to determine this parameter using the experimental maximum pressure values for loading densities of  $0.1$  and  $0.2 \text{ g/cm}^3$ . These values were determined with some measurement uncertainty. The reason for the largest differences in the values of the covolume is additionally the fact that the constancy of the covolume was assumed in the method of its determination based on the measured maximum pressures. Meanwhile, the thermochemical calculations show that the covolume values depend on the loading density. Figure 5 shows, for example, the dependence of the covolume on the loading density for propellants P1, P6 and JA2.



**Figure 5.** Dependence of the covolume of the gaseous combustion products on the loading density for selected propellants

Based on the analysis of the results in Table 6 and the curves shown in Figures 2-4, it may be concluded that the P5 and P6 propellants have ballistic characteristics most similar to those of JA2. However, their vivacities are much smaller. Moreover, their heats of combustion are slightly lower. Nevertheless, these are very promising propellants from the point of view of use in ammunition.

## 5 Summary

In the present work, the heat of combustion was determined in a calorimeter and the pressure histories were measured in a manometric bomb for new RDX-based propellants. Such ballistic parameters as maximum pressure, propellant force and covolume of gaseous products were determined based on the experimental results and calculation using the CHEETAH code. The vivacity was determined on the basis of the pressure histories. The sensitivities to friction and impact were also determined for the tested propellants.

A comparison of the experimental and calculated parameters show that there is quite good agreement between them. The results also indicate that some of the RDX-based propellants are characterized by ballistic parameters comparable to those of JA2. Moreover, the tested propellants are less sensitive to impact and friction than the JA2 propellant. Some of the tested RDX-based propellants may be used in ammunition.

## Acknowledgments

This paper contains the results of investigations carried out under the project co-financed by the National Center for Research and Development in the years 2016-2019 under the terms of Project No. DOB-BIO8/05/01/2016.

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Received: October 31, 2019

Revised: June 11, 2020

First published online: June 25, 2020