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EXPERIMENTAL MEASUREMENTS OF BURNING PARAMETERS OF SELECTED ENERGETIC COMPOSITIONS BASED ON ALUMINUM, BORON AND MAGNESIUM POWDERS AND ALLOYS

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The aim of this study is to determine the relationship between burning rate and flame temperature of various pyrotechnic formulations based on perchlorate, paraffin and metallic powders, and certain SEM-EDS observable physical variables such as mean dimension, morphology and the oxide layer thickness of the metallic powders used as fuel components within a variety of energetic materials.

The comparative analysis indicates a relationship between the burning rate and the mentioned physical characteristics, while the flame temperature is influenced by the chemical compositions of powder or alloy used in development of energetic mixtures. The selected candidates for analysis, aluminum, magnalium (Mg-Al alloy), boron powder and MgB_2, having different particle dimensions and morphologies, were used in mixtures alongside oxidizer (potassium perchlorate) and binder (wax).

The burning rate and flame temperature were determined using ultrafast infrared camera. The results indicate and increase in bu rate with smaller particles in the range 0.75-1.75 mm/s. An interesting speed of combustion was observed for the composition based on boron in comparison with aluminum-based mixture. The measured burn rate is almost ten times higher in the former formulation. The obtained results constitute an perfect background within the selecting process of suitable fuel components in composite energetic materials which have polymeric binders and which can influence the burning rate.

Keywords: aluminum, boron, burning rate, flame temperature, magnesium

1. Introduction

Metallic powders such as magnesium (Mg), aluminum (Al) and boron (B) are widely used as fuels in the development of energetic mixtures due to their exothermic behavior. Their high calorific values increase the flame temperature. Experimental investigations indicate that low boiling values allow excess metal powder in the mixture to vaporize and burn with oxygen in the air, providing

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additional heat [1]. The low cost and relative high yield efficiency can be found in the case of Al which is characterized for its extremely combustible or pyrophoric nature. The performance of pyrotechnic formulations based on metallic powders depend on a number of factors which include their purity, proportions, particle size and morphology, formulation density, mixture blending uniformity, moisture absorbed during development and oxide layer thickness [2]. Ballistic parameters such as burning rate and flame temperature are indicators for selecting the suitable components. Burning rate and flame temperature of metallic powder-based energetic materials have been reported by previous studies that analyzed the ignition and combustion of MgB_2 under atmospheric conditions [3]. Results indicated a combustion temperature of approximately 2000K. The investigations of the properties and dynamics of combustion of boron particles under various atmospheric conditions concluded that the temperature of amorphous boron in air, oxygen and CO_2 is respectively 710 ℃, 697 ℃ and 770 ℃ [4]. Merzhanov et al. [5] reported a combustion temperature of 2300K and an activation energy of 17 kcal/mol. Breiter et al. [6] studied the influence of the oxide layer on the ignition of aluminum particle. Ermakov et al. [7] experimentally determined a temperature between 2000-2100K for aluminum combustion. Anatoly P Denisyuk et al. [8] indicates that at low pressure, Al-Mg alloy significantly increases the burning rate of compositions in comparison with Al.

In this study, pyrotechnic compositions consisting in various metallic powders (aluminum, boron, MgAl and MgB_2), oxidizer (potassium perchlorate) and binder (wax) were developed and experimental analyzed to determine a relationship between the particle size, morphology, oxide layer thickness and combustion parameters.

2. Experimental section

2.1. Materials

Four categories of metallic fuels: aluminum (Al, spheroidal powder, particle size <45 µm, purity >97%, Pyrogarage, Poland), magnalium (Mg-Al alloy 1:1 weight ratio, amorphous powder, particle size <63 µm, Pyrogarage, Poland), boron (B, amorphous powder, average particle size 1-5 µm, purity >95%, dark brown Tanyum, China) and boron-magnesium alloy (MgB_2, amorphous powder, average particle size 1-5 µm, purity >95%, dark brown Tanyum, China) were involved in the development of pyrotechnic compositions as received. Potassium perchlorate ($KClO_4$, purity >98%, Sigma Aldrich), paraffin wax (purity >98%, Sigma Aldrich), toluene (anhydrous, purity >99,8%, Sigma Aldrich) and benzene (anhydrous, purity >99,8%, Sigma Aldrich) were used without further chemical interventions.

2.2 Methods

Pyrotechnic compositions development

Given the heterogeneous structure of an pyrotechnic mixture: oxidizer, metallic fuel and binder, the development process was performed through the following working directions:

The first step consisted in obtaining the solid mixture consisting of oxidizer and metallic powder. Therefore, the oxidizer was sieved, dried and dosed, and mixed with the fuel until a homogeneous distribution was observed, to be further used in energetic formulations. The second step involved preparation of binder by hot dissolution of paraffin in toluene. The paraffin solution was added over the mixture of solids (oxidizer-metallic fuel) and mixed until complete homogenization, followed by evaporation of the toluene from the composition by drying and venting at 40 ℃ for 24 hours. Table 1 depicts the composition of the pyrotechnic formulations.

Table 1

The developed pyrotechnic formulations

Pyrotechnic acronym	Compound proportion [%]					
	Metallic fuel				$KClO_4$	Paraffin wax
	Al	MgAl	B	MgB_2		
CP-Al	20				70	10
CP-MgAl		20			70	10
CP-B			20		70	10
CP-MgB_2				20	70	10

2.3. Characterization

SEM - EDS analysis was performed using a Tescan Vega LMU II microscope coupled to Bruker X-Flash 6 X-ray detector, using advanced vacuum mode, W filament supply of 1.64 kV, 10 kV or 30 kV; the electron gun was focused on the resolution mode using the secondary electron detector (SE - Garnet Yttrium aluminum YAG) as well as the X-ray detector. The samples were analyzed by resolution images at progressive magnification, aiming to obtain images with the distribution and size of the powder, but also detailed images of the surface morphology of the analyzed powders. Measurements were made and an average particle size was determined. In EDS analysis, measurements were performed at a magnification of ≈1000X to obtain an average composition for a representative amount of particles [9]. The spectra represent the average of the measurements made during 10 minutes, at a frequency of 10 scans per second [10]. The carbon signal was excluded from the quantitative calculations, its presence in the spectrum being due to the substrate of the powder fixation band.

Tests on the determination of burning rate and flame temperature were performed using an experimental set-up consisting of a sample holder, a firing system and two acquisition systems, visible spectrum video and thermal imaging. Recording of the visible spectrum was done with a Nikon D3300 camera, with a 52mm, 18-55mm, 1: 3.5-5.6G VR II Nikon AF-S DX lens, focal distance from 0.28m to ∞. The record acquisition rate was 59.94 frames per second. Recording in the thermal spectrum was done with a FLIR X6580sc camera, with 50mm 2.0 640x51 MW lens, with NA_3970-4010 filter, calibrated for the 300-1500 °Crange, the acquisition frequency being 200 frames per second, integration time 30 µs, emissivity set to 0.75 [11], distance between objective and object of 3.5 m, reflected temperature 15 °C, atmospheric temperature 10 °C, relative humidity 30%, atmospheric transmittance factor 1.

3. Results and discussion

3.1. Metallic fuels morphological and chemical characterization

Magnalium has been used in mixtures to study the opportunity to optimize energetic performance by using a powder alloy that combines the high calorific value of aluminum with the low ignition temperature of magnesium. Based on the data reported in literature [12, 13, 14], boron is considered to be a powder which possess the highest mass and volumetric energy density of all metallic powders tested (aluminum, magnesium, titanium, zirconium, carbon, etc.). However, the difficult initiation caused by the oxide layer that forms on the surface of the particles, which has a very high melting temperature, is one of the disadvantages of boron. For this reason, both boron powder and its alloy with magnesium have been studied in this paper.

Sample preparation

To perform the analyses, metal powder samples (MgAl, Al, B and MgB2) were prepared in amounts of 250 mg, dried for two hours in an oven at 40 °C and dispersed in benzene. An amount of approximately 0.3 ml of the suspended powder was taken under stirring with a pipette and dispersed on the surface of a microscope slide. After evaporation of the solvent by drying in an oven, the samples were transferred to an SEM sample holder, on the surface of which carbon tape was fixed. The samples were purged with compressed air and placed under the microscope. The preparation procedure is depicted in Fig. 1.

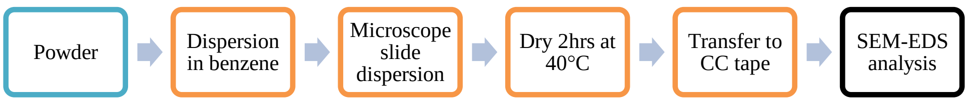

Fig. 1. Sample preparation procedure for the SEM-EDS analysis

Aluminum characterization

From the analysis of the atomized aluminum sample (Fig.2), it can be seen that it has a spheroidal shape, probably obtained by electric arc melting or spraying in an inert atmosphere.

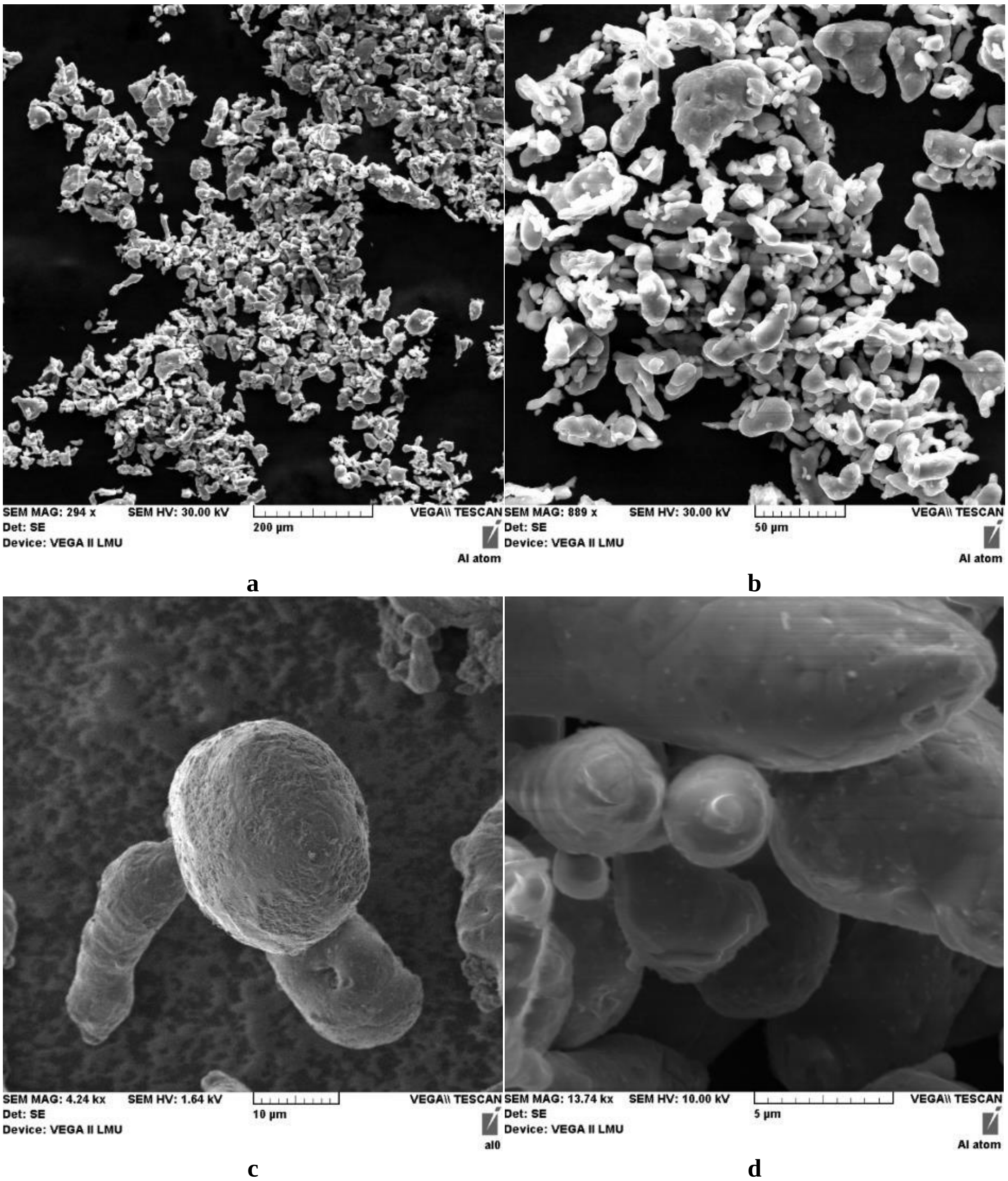

Fig. 2. SEM characterization of the atomized aluminum powder

The particle size varies from approx. 5 µm to dimensions of about 100 µm, with a majority fraction below 15 µm. The low voltage analysis (1.64 kV) allowed the observation of the morphology of the outer oxide layer (see Fig. 2.c), in the form of irregular surfaces, with ribs, as a result of the contraction process at solidification.

The EDS analysis indicated a high degree of purity of the powder, as no chemical elements other than aluminum and oxygen were detected on its surface. The carbon signal comes from the carbon tape on which the powder sample was attached. It can be seen in Fig. 3 how the oxygen signal is more intense in areas where there are irregularities in the shape of the particle, the morphology of these areas causing a higher mass presence of the oxide layer, compared to smooth areas. Preliminary surface analysis indicates a concentration of 1.68% oxygen.

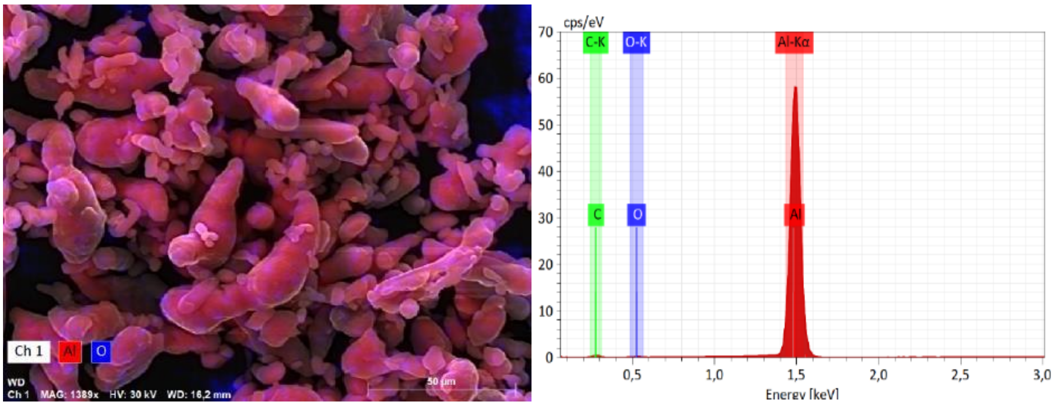

Fig. 3. EDS characterization of the atomized aluminum powder

Magnalium characterization

Magnalium powder is in the form of irregular particles, with sharp edges, being composed of particles with a size between 50 µm and 300 µm, with a majority fraction between 70-120 µm (see Fig. 4). There are two types of powders, on the one hand, large particles with regular surfaces, and on the other hand particles that seem to be obtained by mechanical alloying of several particles with smaller dimensions. Considering the appearance, it can be assumed that the powder was obtained following an abrasion process on a piece of Al-Mg alloy.

From an elemental point of view, it is not possible to distinguish areas with predominant aluminum and areas with predominant magnesium, which leads to the idea that the material from which the powder was made was obtained from melting Al and Mg in an inert atmosphere and not by mechanical alloying of Al and Mg powders. The chemical composition on the surface of the analyzed powder indicates an approximately equal concentration of Al and Mg (49.14% Mg and 44.69% Al by mass), while the concentration of oxygen (6.17% by mass) indicates the existence of a very thin layer of oxide.

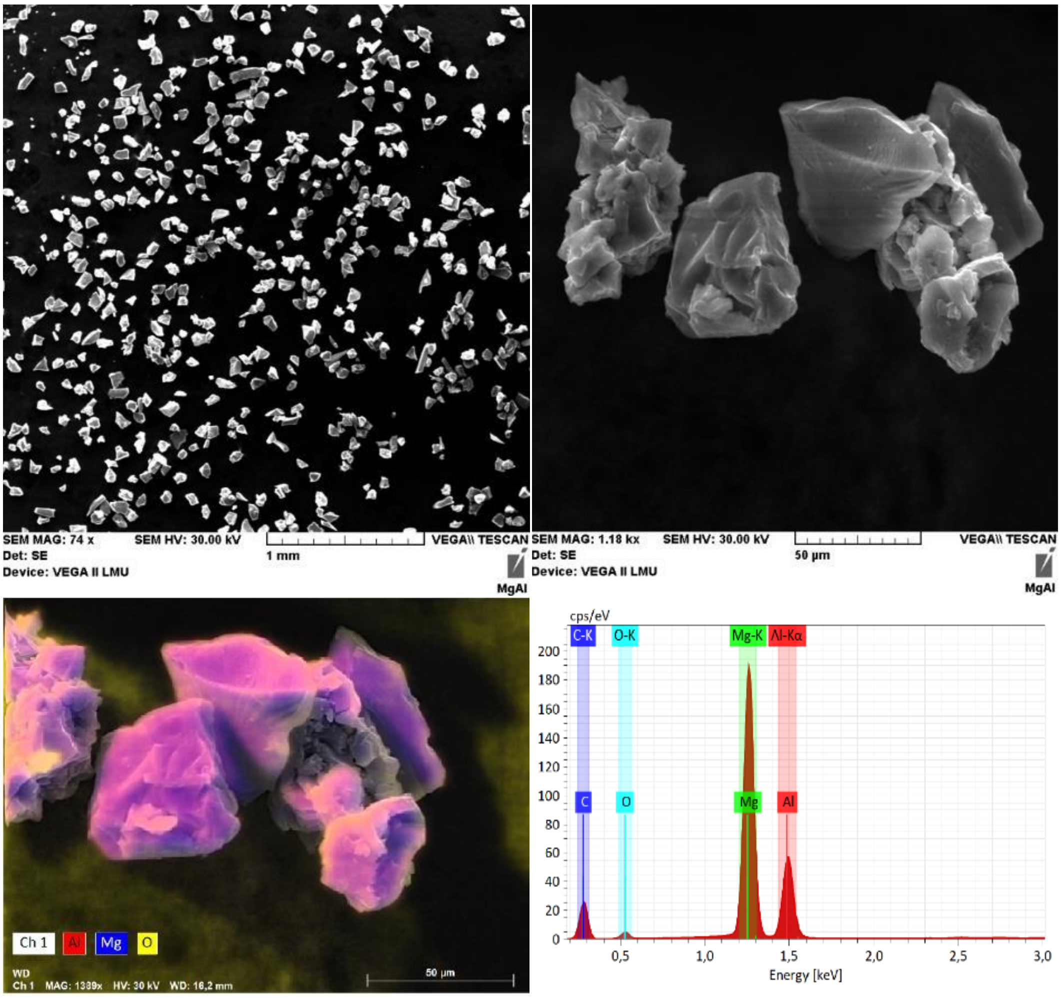

Fig. 4. SEM-EDS analysis of magnalium powder

Boron characterization

Boron powder is in the form of flakes with irregular geometry, being agglomerated in large clusters (30-70 µm). The particles are presented in the form of plates with widths in the range of 2-5 µm and thickness in the range of 100 nm-1 µm (Figure 5). These particles have a special morphology, with dendrites at the edge. This plate morphology with dendritic edges gives the powder a very large specific surface area.

The elemental composition on the surface of the powder (considering the very small thickness of the particles, this composition can be considered as representative for the whole substance) is composed of boron (96.23% by weight) oxygen (3.01% by weight) and traces of fluorine, magnesium, calcium, potassium and silicon (0.37%, 0.12%, 0.10%, 0.10% and 0.07% by weight). The low concentration of oxygen indicates the existence of a very thin layer of oxide, which leads to a reduced weight fraction of boron oxide, even if the very large specific surface area of the powder could favor a sustained oxidation.

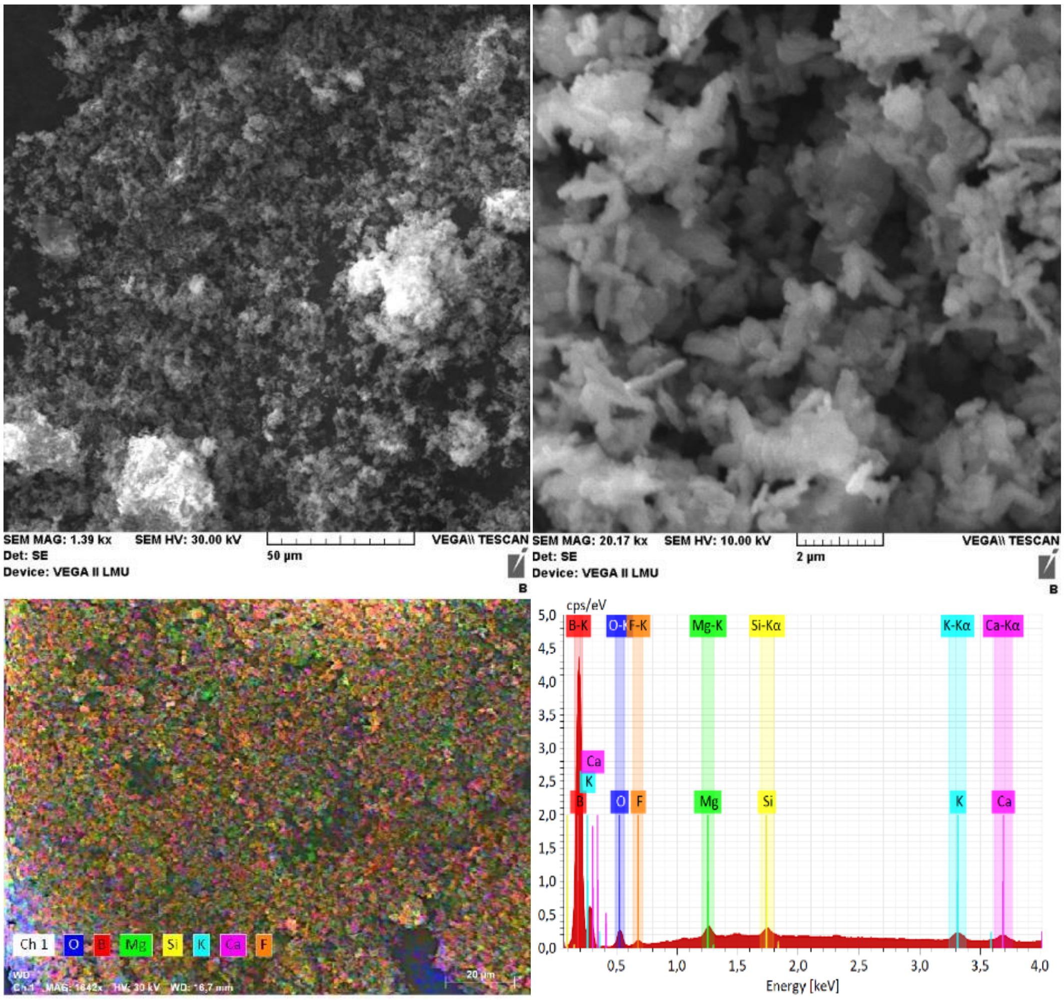

Fig. 5. SEM-EDS analysis of boron powder

Magnesium-boron characterization

The powder is in the form of irregularly shaped flakes, with rounded edges, with a wide distribution of particle sizes (5-150 µm). The average particle size is around 10 µm. Two types of particles can be observed, some larger (≥30 µm) with regular morphological appearance (spheroidal) and regular surface and some small particles (5-10 µm) with irregular surface, as is illustrated in Fig. 6. The chemical structure on the surface indicates distinct areas in the composition of the powder. The larger particles being composed mostly of magnesium, while the smaller particles contain boron and oxygen. This indicates a mechanical alloying of the small powder (boron) on the surface of the larger powder (magnesium). It is observed that the material is not homogeneous and the functional role of boron alloying with magnesium is not achieved, due to the fact that magnesium is not distributed on the surface of boron particles, but vice versa, practically no piercing of the boron oxide layer taking place. Moreover, it is very likely that due to the difference in density and particle size, the separation of the two component powders will occur when formulating the explosive mixtures.

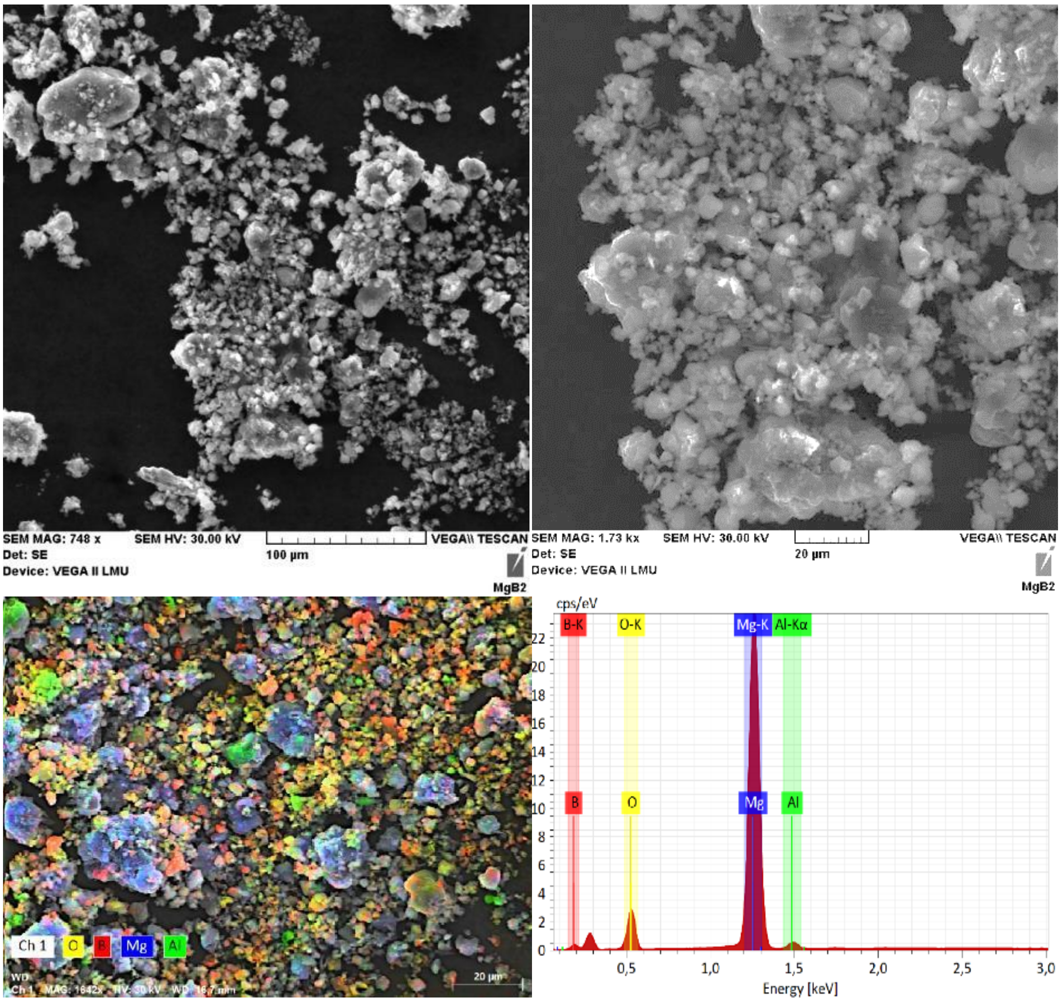

Fig. 6. SEM-EDS analysis of MgB_2 powder

Oxide layer thickness analysis

Based on the assumption that the penetration depth of the electron beam is 2 μm (1-3 μm in the literature) the thickness of the oxide layer can be estimated at the aluminum particles, for each of the samples analyzed as follows, starting from several simplifying hypotheses [15]:

- the surface is covered only with oxide;
- the surface is spherical and smooth in appearance;
- the average depth from which the X-ray signal comes is maximum 2 µm.

The results for the elemental analyses are briefly presented in Table 2, while the estimates for the thickness of the oxide layer are presented in Table 3.

Table 2

Elemental analysis of fuel powder surface

Chemical compound / Metallic fuel	Al		O		Mg		B		Si		Other chemical compound		
	Weight (%)	Atomic (%)	Weight (%)	Atomic (%)	Weight (%)	Atomic (%)	Weight (%)	Atomic (%)	Weight (%)	Atomic (%)	Weight (%)	Atomic (%)	Element symbol (impurity)
Atomized aluminum	98.32	97.20	1.68	2.80									
Mg-Al	44.69	40.76	6.17	9.48	49.14	49.76							
Boron powder			3.01	2.06	0.12	0.06	96.23	97.59	0.07	0.03	0.37	0.21	F
											0.1	0.03	Ca
											0.1	0.03	K
MgB_2 alloy	0.79	0.41	24.09	20.87	24.72	14.10	50.40	64.63					

Table 3

Estimation amount and thickness of the oxide layer for metallic powders

Sample	Al total [%] weight	O [%] weight	Oxide [%] weight at the exterior	Metallic Al [%] weight at the exterior	Oxide layer thickness [nm]	Oxide fraction in powder [%] weight
Atomized aluminum	98.32	1.68	2.6	97.4	36	0.59*
Boron powder	97.00	3	3.7	96.3	50	3.7

** Estimated for a spherical oxide particle with a diameter of 50 µm.*

In the case of relatively large particles (≥10 µm) the elemental analysis is representative of the outer area of the particle. In this case, the mass percentage of oxide in the powder is lower (respectively for the atomized aluminum powder), the purity of these powders being estimated (considering the spherical particle with a diameter of 50 µm) to 99.4% for the atomized aluminum powder. In the case of boron powder, the elemental composition determined by EDS is representative of the entire amount of powder.

As can be seen, the smaller the powder size, the higher the mass percentage of the oxide layer and, implicitly, the powder will have a lower energy content. On the other hand, combustion a powder with smaller particles will increase the burning rate, the large amount of energy being released in a shorter period. Regarding the thickness of the oxide layer, it is observed that the atomized powder has the best ratio between the thickness of the oxide layer and the amount of oxidizable aluminum in its composition. The thinner the oxide layer, the easier the powder is expected to ignite.

3.1. Metallic fuels morphological and chemical characterization

To investigate the combustion of metallic powders used in pyrotechnic mixtures, experiments were performed under isobaric conditions (atmospheric pressure). The thermochemical properties of the reactants used in the pyrotechnic mixtures are shown in Table 4. The composition of the pyrotechnic mixtures and the characteristics of the mixtures made are described in Table 5.

Table 4

Thermochemical characteristics of components used into pyrotechnic compositions

Component	Equivalent chemical formula	Maximum theoretical density [g/cm^3]	Heat of formation [kJ/mol]	Reference
Potassium perchlorate	$KClO_4$	2.528	-432	[16]
Paraffin	$C_{7.12}H_{14.4}$	1.09	-127.83	[16]
Aluminum	Al	2.699	0	[16]
Amorphous boron	B	2.37	0	[16]
Magnalium	MgAl	2.086	- 2,2	[17]
MgB_2	MgB_2	2.113	-91,96	[18]

The burning rate was measured for cylindrical grain (diameter of 13 mm), by pressing a quantity of approximately 3 g of pyrotechnic composition, at a pressure of 1.27 MPa. To ensure a proper combustion the grain cylindrical surface was inhibited with self-adhesive aluminum tape (see Fig. 7).

Table 5

The characteristics of pyrotechnic compositions

Sample	CP-Al	CP-MgAl	CP-B	CP-MgB_2
Reactant heat of formation [kJ/kg]	-2310.38			
Oxygen balance [%]	-19.74	-17.66	-46.35	-22.95
Maximum theoretical density [g/cm^3]	2.258	2.176	2.207	2.158
Obtained density [g/cm^3]	2.068±0.075	1.829±0.009	1.735±0.007	1.898±0.035
Pellet height [mm]	10±0.35	11.3	12.09±0.01	10.89±0.05
Sample weight [g]	2.912±0.043	2.913±0.015	2.955±0.015	2.914±0.062

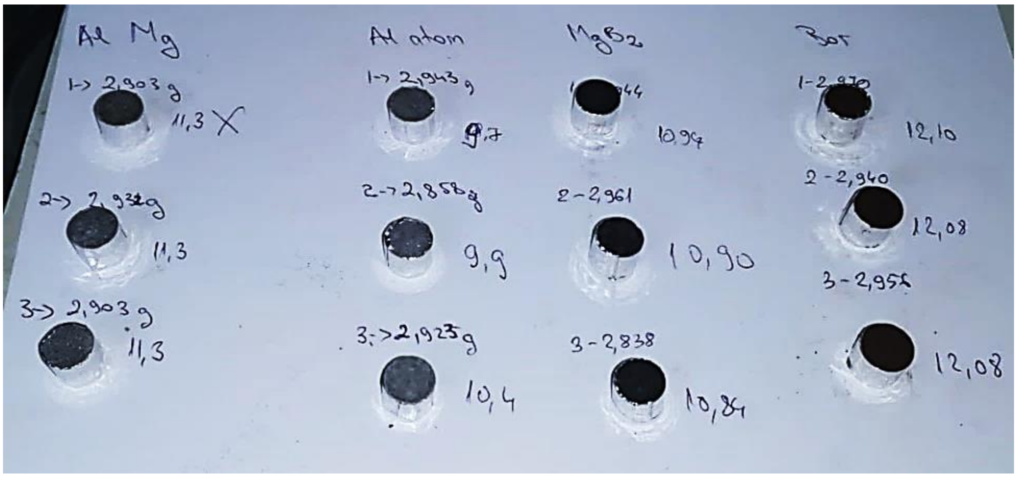

Fig. 7. Cylindrical grain pyrotechnic compositions molds

In Fig. 8 is presented the set-up for measuring the burning rate of the developed pyrotechnic formulations.

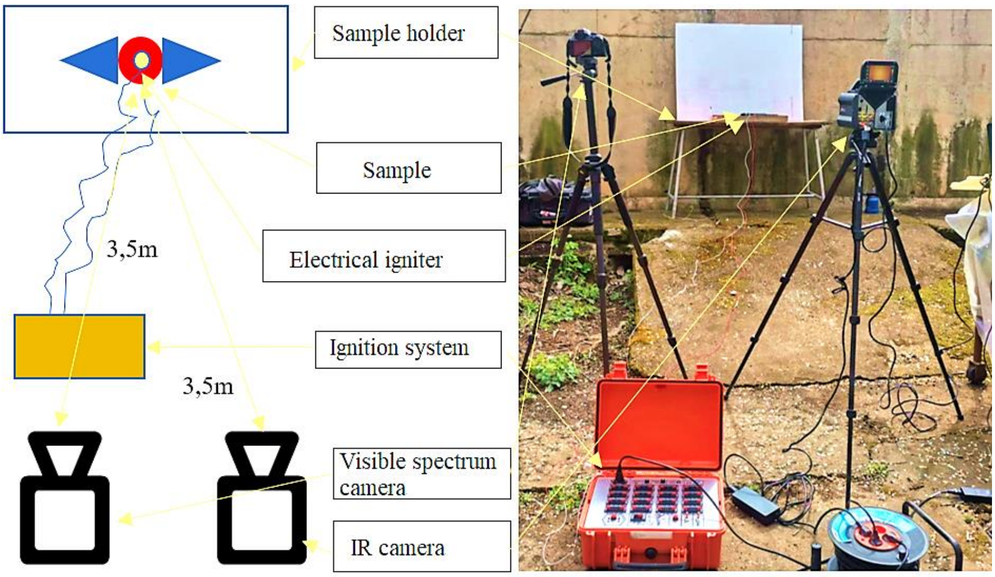

Fig. 8. Setup for measuring the rate of combustion

The ignition of the compositions was performed with an electric igniter of 0.3 g (ignition composition based on potassium chlorate and lactose) and 0.1 mm nickel filament initiated with a firing system that provides a maximum current of 3A, at a voltage of 12V, for 1s. The compositions CP-Al, CP-MgAl and CP-MgB_2 were not ignited by this method nor by the addition of a powdered primer (0.3 g) which were initiated with the gas lamp (prolonged exposure until self-ignition). The

CP-B composition was initiated with the electric igniter. Fig. 9 shows the maximum temperature variation (maximum value of all acquisition points 640x480 pixels) - combustion time for each pyrotechnic composition. Fig. 10 displays representative frames from the acquisitions for the tested mixtures.

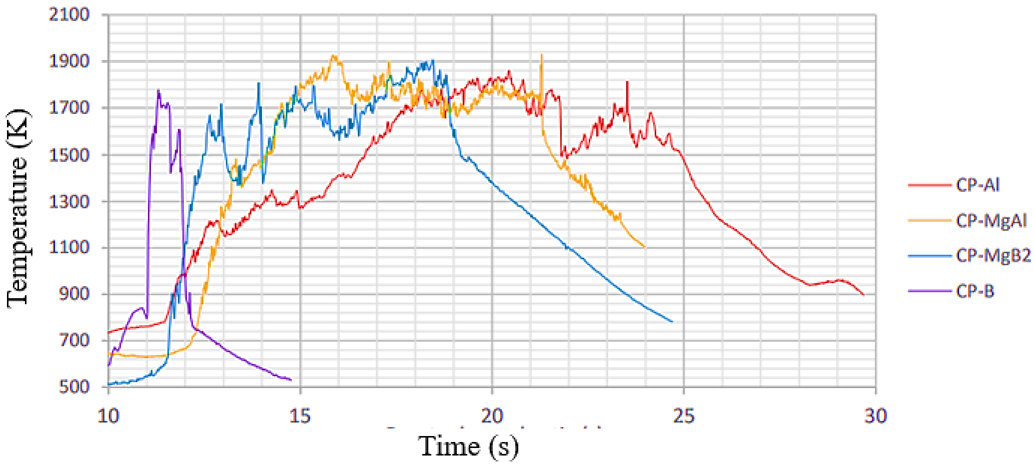

Fig. 9. Temperature variation as function of combustion time

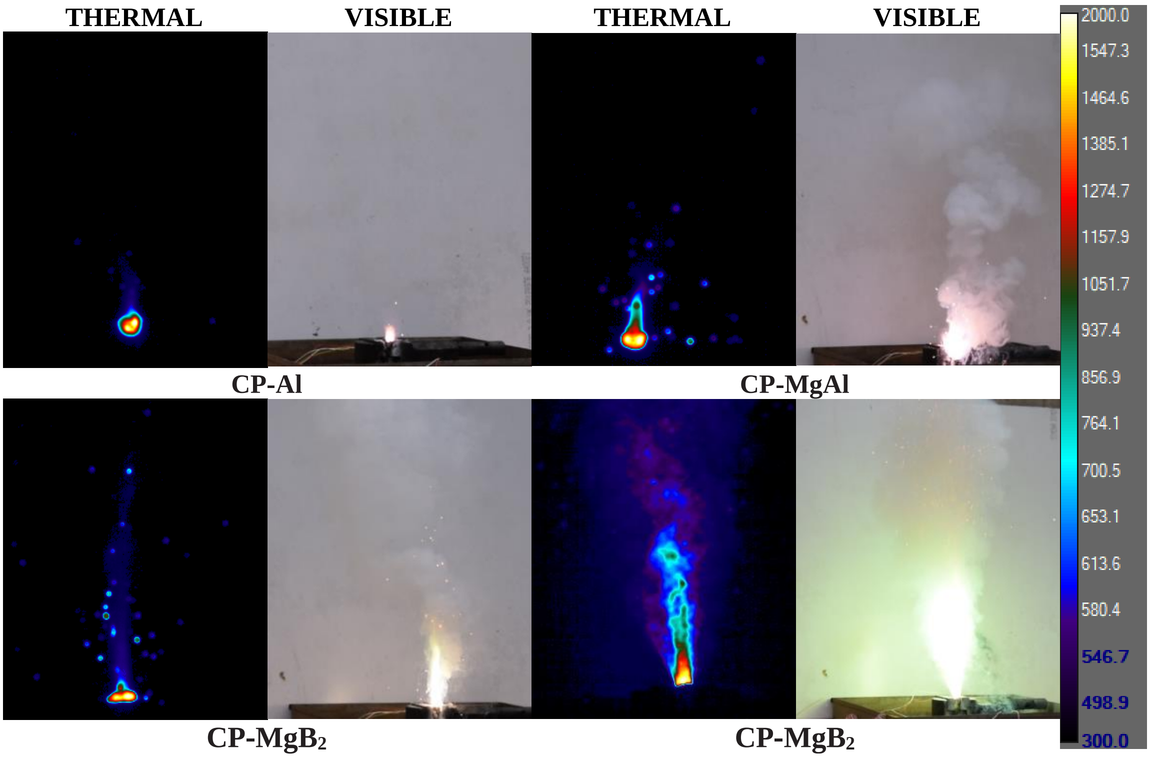

Fig. 10. Flame configuration for pyrotechnic formulations

Based on data depicted in Table 6, it can be seen that the measured burning rate is in the range of 0.75-1.75 mm/s for the pyrotechnic mixtures CP-Al, CP-MgAl, CP-MgB_2, except CP-B which is approximately 14mm/s, while the maximum and maximum time-averaged temperature are placed in the range 1800–1950 K and 1500–1650 K, respectively.

Table 6

Temperature and rate of combustion of metal powder compositions

Sample	CP-Al	CP-MgAl	CP-MgB_2	CP-B
Combustion rate [mm/s]	0.75±0.03	1.29±0.04	1.44±0.06	13.44±0.73
Maximum absolute temperature [K]	1937±57	1860±69	1942±23	1776±81
Time average maximum temperature [K]	1507±14	1615±34	1657±64	1572±32

4. Conclusions

In this study, the effects of particle size, morphology and oxide layer thickness of different metallic powders on the combustion behavior of pyrotechnic compositions were investigated. The results observed were more than satisfactory. So, it seems that the pyrotechnic composition based on boron powder has a special behavior, this manifesting a higher burning rate (≈14 mm/s - almost ten times higher than the speed combustion of CP-Al). This peculiarity can be explained both by the very large specific surface area of the boron powder and the small size of particles, which leads to the existence of a much larger combustion surface, as well as due to the lower loading density achieved for the same applied pressing force. The morphological appearance of the powder is not sufficient to explain the high combustion rate, as CP-MgB2 also has a double combustion rate compared to CP-Al, although both the specific surface area and the particle size of magnesium diboride are more disadvantageous in terms of combustion rate compared to aluminum. The explanation of the net higher combustion rate of the boron powder composition depends on both the morphology of the powder and the thermochemical properties of boron and, in particular, of the oxide layer (its melting).

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R E F E R E N C E S

[1]. *Agrawal J.P.*, High energy Materials, Propellants, Explosives and Pyrotechnics. J. Wiley-VCH, Verlag GmbH & Co., KGaA, Weinheim 2010.

[2]. *Koch E.-C.*, Metal-Fluorocarbon Based Energetic Materials, John Wiley & Sons, 2012, pp. 104-105.

[3]. *Pang, W., De Luca, L.T., Fan, X., Glotov, O.G., & Zhao, F.* (2019). Boron-Based Fuel-Rich Propellant: Properties, Combustion, and Technology Aspects (1st ed.). CRC Press.

[4]. *A. G. Merzhanov, Y.. M. Grigorjev, Y. A. Gal'chenko*, Aluminum ignition, Combust. Flame, **vol. 29**, 1977, pp. 1-14.

[5]. *A. L., Breiter, V. M. Mal'tsev, E. I. Popov*, Models of metal ignition, CESW, **vol. 13**, no. 4, 1977, pp. 475-484.

[6]. *V. A. Ermakov, A. A. Razdobree A. I. Skori, V. V. Pozdeev, S. S. Smolyakov*, Temperature of aluminum particles at the time of ignition and combustion, CESW, **vol. 18**, no. 2, 1982, 256-257.

[7]. *A. P. Denisyuk, N. D. Tuan,V. A. Sizov*, Combustion Behavior of the Inorganic Nitrates-Based Compositions. Part II: Effect of Al and Al-Mg Alloy on Burning Rate, Propellants, Explosives, Pyrotechnics, **vol. 45**, 2022.

[8]. *Türker L.*, Thermobaric and enhanced blast explosives (TBX and EBX), Defence Technology, **vol. 12**, 2016, pp. 423-445.

[9]. *Wanjun Zhao, Qingjie Jiao, Pengwan Chen, Shi Yan, Yanli Zhu, Bi Zhang, Xin Zeng, Dazhi Liu, Yapeng Ou, Fang Wang*, Synergetic energetic kinetics of Mg-Zn alloys and pyrotechnics,Combustion and Flame, **vol. 240**, 2022, 112000, ISSN 0010-2180.

[10]. *X.-H. Li, F. Zhao, J.-C. Qin, H.-B. Pei*, A new method for predicting the detonation velocity of explosives with micrometer aluminum powders, Propel. Explos. Pyrotech., **vol. 43**, 2018, 333.

[11]. *C. Lăzăroaie, S. Eşanu, C. Său, R. Petre, P. Z. Iordache G. Staikos, T. Zecheru,* Temperature measurements of magnesium-and aluminum-based flares, J. Therm. Anal. Calorim, **vol. 115**, no. 2, 2013, pp. 1407-1415.

[12]. *W.A. Trzcinski, L. Maiz*, Thermobaric and enhanced blast explosives – properties and testing methods, Propellants Explos. Pyrotech., **vol. 35**, 2010, pp. 1-14.

[13]. *C. Ludwig*, Verifying performance of thermobaric materials for small to medium caliber rocket warheads, 38th Annual Guns, Ammunition & Missiles, Symposium & Exhibition, Arlington, Virginia, SUA, 2003.

[14]. *Erwin Vermeij, Willem Duvalois, Rutger Webb, Mattijs Koeberg*, Morphology and composition of pyrotechnic residues formed at different levels of confinement, Forensic Science International, **vol. 186**, no. 1–3, 2009, Pages 68-74, ISSN 0379-0738.

[15]. M. Sućeska, Explo5 User Guide, EXPLO5, Version 6.05 /Edition 1/2018.

[16]. *C. Epure, T.-V. Tiganescu, O. Iorga, A. Marin, M. Munteanu, T. Zecheru, A. Schiopu, B. Pulpea*, Fuel ratio and additives influence on the combustion parameters of novel polyurethane-based flares, Mater. Plast., **vol. 57**, no. 4, 2020, pp. 109-124.

[17]. Shah, Luqman & Gerlich, A. & Yushan, Zhou. (2018). Design guideline for intermetallic compound mitigation in Al-Mg dissimilar welding through addition of interlayer. The International Journal of Advanced Manufacturing Technology. 94. 10.1007/s00170-017-1038-y

[18]. Chase, M.W., Jr., NIST-JANAF Themochemical Tables, Fourth Edition, J. Phys. Chem. Ref. Data, Monograph 9, 1998, 1-1951