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# Certain Ballistic Performance and Thermal Properties Evaluation for Extruded Modified Double-base Propellants

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**Abstract:** The main advantages of modified double-base (MDB) propellants are wide range of burning rates, high energy output, as well as enhanced thrust. This study reports on the effect of potential oxidizers − potassium perchlorate (KP) or ammonium perchlorate (AP), stoichiometric binary mixture of the oxidizer (KP or AP) with metal fuel (Al), and energetic nitramine (HMX) on combustion characteristics of MDB propellants. MDB propellant formulations based on these additives, constituting 10 wt.% of total mass of the MDB formulation, were manufactured by solventless extrusion process. The impact of these additives on ballistic performance particularly the burning rate as well as on the characteristic exhaust velocity of gaseous product ($C^*$), was evaluated using small-scale ballistic evaluation test motor. KP and AP exhibit different effects; KP positively impacts the burning rate, AP positively impacts $C^*$. Stoichiometric binary mixture of AP/Al positively impacts both the burning rate and $C^*$; HMX substantially enhances $C^*$. These energetic additives could alter the combustion mechanism, by thinning the induction zone, allowing the luminous flame zone to be more adjacent to the burning surface. Therefore, the combustion reaction could proceed faster. The developed MDB propellant formulations were found to be more energetic with an increase in calorific value in comparison to reference formulation (using bomb calorimeter); they exhibited similar ignition temperature by means of cook off test. DSC measurements demonstrated similar onset and maximum decomposition temperature of developed MDB propellant formulations to reference DB propellant formulation but with an increase in total heat released (J/g).

**Keywords:** solid propellant, modified double-base propellant, combustion, burning rate, ballistic performance, thermal properties

## 1 Introduction

It is widely accepted that modified double-base (MDB) propellants are evolved from double-base propellants by introduction of energetic fillers such as HMX or RDX [1-3]. There is also another trend to introduce potential oxidizers such as ammonium perchlorate (AP) or potassium perchlorate (KP) as well as active metal fuels such as aluminum, magnesium, and boron into MDB propellants [2, 4, 5]. Consequently, MDB propellants can offer wide range of burning rate and specific impulse [6]; this is why MDB propellants have recently been used in booster, sustainer, and dual thrust rocket motors [7]. MDB propellants have wide applications in tactical missiles due to their advantages including superior mechanical properties, aging capabilities, and good operational characteristics [1, 2, 8, 9]. One of the main operational (ballistic) characteristics of extruded double base propellants, is their high specific impulse and burning rate which could be up to 220 s and 40 mm/s, respectively [10, 11]. Furthermore, the extruded DB propellants can be assembled in rocket motor as free standing grains; thus high burning surface area can be achieved [12]. The burning rate and the specific impulse are the main parameters which need to be precisely optimized and evaluated during combustion process to meet specific propulsion requirements (booster, sustainer, or dual thrust rocket motor) [13].

High reaction rates might be achieved by increasing the combustion temperature, thermal conductivity, an/or operating pressure inside the rocket motor [14-16]. Combustion temperature or pressure could be increased by introduction of different potential oxidizers (KP or AP), active metal fuel (aluminum, magnesium, or boron), or energetic nitramines (RDX or HMX) into DB propellant formulations. These energetic additives have the potential to induce exothermic reactions at higher rate in the induction zone, decreasing its thickness, and hence could increase the burning rate [17]. Much research has been directed toward the development of cast modified double-base propellant formulations; less attention has been directed to the development of extruded MDB propellants with enhanced ballistic performance [18]. This study is devoted to the development of extruded MDB propellants based on potential oxidizers such as KP or AP, binary oxidizer (KP or AP)/metal fuel mixture, and energetic nitramine (HMX). These potential oxidizers, KP or AP, are candidates and even better than energetic nitramines at solid loadings level up to 50 wt.% of total mass of MDB propellants [6, 19]. KP and AP have the potential to enhance the oxygen balance and to react with inert constituents (plasticizers, stabilizers, and ballistic modifiers). Consequently, they have the potential to enhance the heat of combustion of MDB propellant formulations [18]. One of the main features of KP is its exothermic decomposition with release of high amount (mass percentage) of active oxygen. KP has an oxygen content of 46.2 wt.%; this high content allows KP to be one of the most effective oxidizing agents [20, 21]. Equation 1 shows KP decomposition reaction.

$$KClO_4 \longrightarrow KCl + 2\ O_2 \tag{1}$$

On the other hand, AP has found considerable use in modern solid-fuel rocket propellants particularly composite propellants. AP (with active oxygen content of 34%) undergoes a complex chemical reaction on heating (Equation 2) [22].

$$10\ NH_4ClO_4 \rightarrow 2.5\ Cl_2 + 2N_2O + 2.5\ NOCl + HClO_4 + 1.5\ HCl + \\ + 18.75\ H_2O + 1.75\ N_2 + 6.375\ O_2 \tag{2}$$

The fact that AP generates all gaseous products makes it a valuable oxidizer for MDB propellant formulations [23, 24]. Mixture of AP and potassium chlorate should not be used due to the possible formation of unstable (spontaneously explosive) ammonium chlorate in presence of moisture [25]; this action has been verified by Jain, Mehilal *et al*. 2016 [26], further details are given in the reference [26]. Xiangyu Li *et al*. [27] investigated the thermal decomposition properties of DB propellant formulations based on AP. There was an increase in oxygen balance and heat of combustion with the increase in AP content. Bhat *et al*. [28] conducted a systematic study on the combustion characteristics of MDB propellants based on potential oxidizers such as ammonium nitrate, potassium nitrate, AP, KP, RDX and PETN. While AP and KP increased burning rates, other additives maintained either similar or reduced burning rates. Propellants containing these additives showed marginally higher flame temperature.

Fuels also play an important role in determining the rate of combustion. Metal fuels with high heat of combustion and excellent thermal conductivity, tend to increase the rate of burning. The most commonly used metallic fuel is aluminum (Al). Aluminum is safer than other metallic fuels; during storage of MDB propellants, a protective aluminum oxide layer is formed which protects the inner Al surface from further oxidation [7]. Al particles are able to react not only with free oxygen resulted from oxidizer decomposition but they are also able to react with inert decomposition gaseous products and add much more heat to the combustion process [29]. Metal powders also demonstrated a catalytic effect on AP thermal decomposition [30]. The combination of potential oxidizers (KP or AP) and energetic metal fuel could be vital for MDB propellants to achieve highly energetic formulations with high combustion temperature, stable burning rate, and enhanced specific impulse [15]. Alternatively, HMX one of the most energetic explosive materials, can increase the energy of combustion of MDB propellants. It can induce highly exothermic decomposition reaction, offer large amount of gasses with low molecular weight [31].

MDB propellant formulations based on different potential oxidizers (KP or AP), stoichiometric oxidizer/metal fuel binary mixture (AP/Al), and energetic nitramine (HMX), were developed at 10 wt.% loading level of total mass of given MDB propellant formulation for each type of introduced component. The impact of these energetic constituents on MDB propellants ballistic performance, particularly on the burning rate and $C^*$, was evaluated using small-scale ballistic evaluation test motor. Small-scale ballistic evaluation test motor has been established as the most representative mean to obtain accurate determinations of burning rate, operating pressure, and $C^*$ [11]. The two oxidizers KP and AP exhibited different effect. Whereas KP positively impact the burning rate by 22%, AP positively enhanced $C^*$ by 5.3% due to its gaseous decomposition nature (Equation 2). Stoichiometric binary mixture of AP/Al enhanced both burning rate and $C^*$ by 12% and 10%, respectively. HMX increased $C^*$ by 9.5%. The developed MDB propellant formulations were found to be more energetic with an increase in heat of combustion in comparison to the reference DB propellant formulation using bomb calorimetry; they also exhibited acceptable thermal behavior using DSC and cook off tests. This paper might open the route for the manufacture of MDB propellant formulations with tailored performance in terms of specific impulse and burning rate.

## 2 Theoretical Evaluation of Ballistic Performance

Thermochemical methods have been widely used as a valuable fast tool to predict the combustion characteristics of different solid propellant compositions [32-34]. In this study, the effect of different potential oxidizers (KP or AP), stoichiometric oxidizer/metal fuel mixture (AP/Al), and nitramine (HMX) on the combustion characteristics of extruded DB propellants particularly the specific impulse, and the combustion temperature ($T_c$) were evaluated using a chemical equilibrium computer program named ICT Thermodynamic Code (Institute of Chemical Technology in Germany, virgin 2008). This code is based on the chemical equilibrium and steady-state burning model; which is based on two methods of calculation: frozen equilibrium and shifting equilibrium. The employed frozen equilibrium model is based on the assumption that the composition is invariant (the product composition at the nozzle exit is identical to that in combustion chamber). The performance of MDB propellant formulations, at 10 wt.% of each above solid loading/additives level, was evaluated. The chemical compositions of the investigated MDB propellant formulations are listed in Table 1.

**Table 1.** The chemical composition of the reference DB propellant and developed MDB propellant formulations

| | NC [wt.%] | NG [wt.%] | Stabilizers [wt.%] | Other additives [wt.%] | KP [wt.%] | AP [wt.%] | Al [wt.%] | HMX [wt.%] |
|---|---|---|---|---|---|---|---|---|
| $F_0$ (Reference) | 58.20 | 31.55 | 1.80 | 8.45 | 0 | 0 | 0 | 0 |
| $F_1$ (10 wt.% KP) | 52.68 | 28.10 | 1.63 | 7.59 | 10 | 0 | 0 | 0 |
| $F_2$ (10 wt.% AP) | 52.68 | 28.10 | 1.63 | 7.59 | 0 | 10 | 0 | 0 |
| $F_3$ (7 wt.% AP + 3 wt.% Al) | 52.68 | 28.10 | 1.63 | 7.59 | 0 | 7 | 3 | 0 |
| $F_4$ (10 wt.% HMX) | 52.68 | 28.10 | 1.63 | 7.59 | 0 | 0 | 0 | 10 |

Thermochemical calculations demonstrated an increase in the specific impulse with the increase of oxidizing agent content; particularly it concerns KP and AP. This could be ascribed to the enhanced oxygen balance of MDB propellant formulations ($F_1$, $F_2$), which could result in an increase in the heat of combustion and flame temperature [21, 35]. Stoichiometric binary mixture of oxidizer/metal fuel ($F_3$) and HMX ($F_4$) were also found to have a great impact on the specific impulse of MDB propellant formulations (Figure 1).

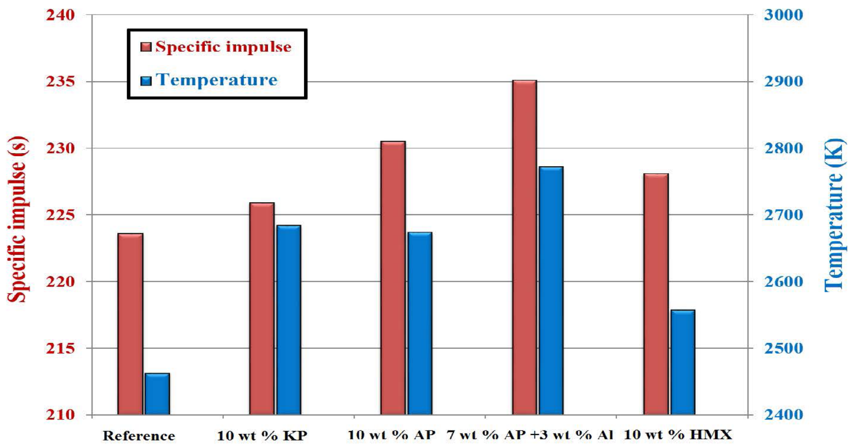


**Figure 1.** Theoretical specific impulse and combustion temperature for DB propellant formulation (reference one) and MDB propellant formulations based on added different energetic materials

Stoichiometric mixture of AP/Al ($F_3$ formulation) demonstrated the greatest impact on specific impulse; this was ascribed to the gaseous decomposition nature of AP and the exothermic oxidation of Al metal fuel which could enhance the combustion flame temperature [7, 36]. Aluminum is an energetic metal fuel which can enhance the heat output due to its high heat of combustion 30.96 kJ/g. Aluminum can also enhance the heat transfer from the burning zone to the adjacent layers of unreacted composition due to its high thermal conductivity [37]. Thus Al has the potential to speed up the burning rate of MDB propellant formulations. On the other hand, HMX as an energetic explosive material with heat of explosion 6.197 kJ/g and large amount of gaseous product of 902 L/kg can exhibit a positive impact on both the specific impulse and combustion temperature [38].

# 3 Experimental

## 3.1 Manufacture of tested MDB propellant formulations

Screw extrusion technique emphasizes mixing of different ingredients to molecular level, good homogenization, high density, and dimensional stability [8]. This technique includes many stages such as blending, followed by rolling, grinding, granulation, and finally extrusion to obtain grains of desired shape and dimensions [39]. Different MDB propellant formulations, based on KP, AP, stoichiometric binary mixture (AP/Al), and HMX (Table 1) were manufactured by screw extrusion. Ultra-fine aluminum in the shape of flakes of 5 μm average particle size was employed in combination with AP. The total solid loading level was limited to 10 wt.% of total mass for each MDB propellant formulation to secure its good homogeneity and integrity (free from defects). For each investigated formulation, a tubular test specimen of accurate dimensions 29 mm OD, 10 mm ID, and 200 mm length was manufactured for ballistic performance evaluation.

## 3.2 Ballistic performance evaluation of DB propellant formulation and MDB propellant formulations

Small-scale ballistic evaluation motor is widely accepted to measure real ballistic characteristics of solid propellants during static firing tests [8, 11]. Ballistic performance of the developed MDB propellant formulations was evaluated using one-inch test motor (33 mm ID and 225 mm length) (Figure 2).

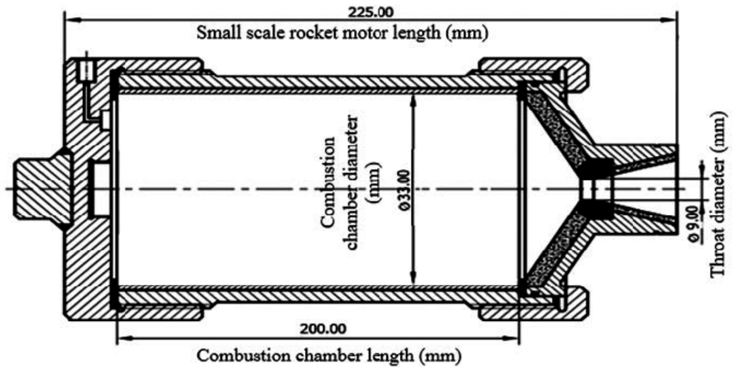


**Figure 2.** Schema for employed small-scale ballistic evaluation test motor

During static firing test, the pressure (p) inside the motor chamber was recorded with time (t) over combustion process (Figure 3). On the basis of p(t) curve, the operating pressure, burning time, burning rate, and characteristic exhaust velocity were evaluated [40]. All MDB propellant formulations exhibited ideal stable burning over the combustion process. This confirmed that tested MDB propellant formulations have good homogeneity, and they were free from air bubbles, cracks, and foreign matter. Whereas, KP considerably decreased the average operating pressure; AP and AP/Al slightly increased the average operating pressure. HMX exhibited similar operating pressure to reference formulation but with extended burning time (Figure 3).

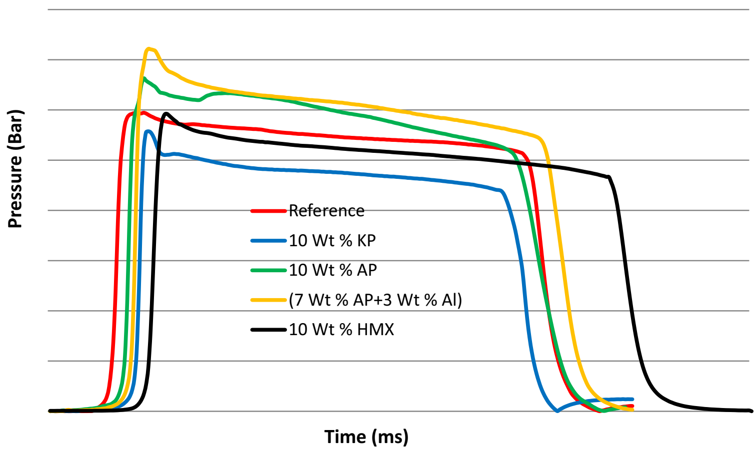


**Figure 3.** Pressure-time (p(t)) curves of reference DB propellant formulation and developed MDB propellant formulations using small-scale ballistic evaluation rocket motor

Characteristic exhaust velocity ($C^*$) is one of the key indicators for the specific impulse. The total area under the P-t curve was computed to retrieve the characteristic exhaust velocity using Equation 3.

$$C^* = \frac{I_{PT} \times A_t}{W_t} \tag{3}$$

Where: $C^*$, $I_{PT}$, $A_t$, $W_t$ are the characteristic exhaust velocity, the total impulse, throat area, and propellant mass respectively. The main combustion characteristic parameters, including $C^*$, burning rate, and average operating pressure of the developed formulations, are presented in Table 2.

**Table 2.** Results on ballistic (performance) parameters of reference DB propellant formulation and MDB developed propellant formulations using the one-inch small-scale ballistic test motor

| | Characteristic exhaust velocity ($C^*$) [m/s] | Burning rate [mm/s] | Average operating pressure ($P_{av}$) [bar] |
|---|---|---|---|
| $F_0$ (Reference) | 1368 | 25.68 | 241.74 |
| $F_1$ (10 wt.% KP) | 1400 | 31.33 | 210 |
| $F_2$ (10 wt.% AP) | 1440 | 26.55 | 257.07 |
| $F_3$ (7 wt.% AP + 3 wt.% Al) | 1505 | 28.57 | 291.48 |
| $F_4$ (10 wt.% HMX) | 1496 | 25.23 | 234.8 |

It is apparent that the main advantage of KP at 10 wt.% is the considerably increase in burning rate by 22%; this was ascribed to the exothermic decomposition nature of KP, as well as to its strong oxidizing power (oxygen content of 46.5%) [7]. In the meantime, MDB propellant formulation based on KP demonstrated the lowest average pressure inside the rocket motor (210 bar); this was correlated to the fact that KP exhibits less gaseous decomposition products as described in Equation 1. The relatively small amount of gaseous decomposition products could cause the decrease in the average operating pressure. The main advantage of AP over KP, is existence of much more gaseous products during AP decomposition than during KP decomposition, this could be reason that AP positively impacts the average operating pressure and $C^*$.

Aluminum metal fuel also plays an important role in determining the burning rate of combustion process. Metal fuels, with high exothermic heats of combustion and excellent thermal conductivity values, have the potential to increase the burning rate [7]. This why binary mixture of AP/Al exhibited higher burning rate compared to MDB propellant formulation based on AP alone; the high heat of combustion due to reactive metal oxidation could be reason of the highest value of pressure value inside the rocket motor as well as the highest $C^*$. Aluminum particles are able to react not only with free oxygen resulted from oxidizer decomposition but they are also able to react with inert decomposition gaseous products and add much more heat to the combustion process [41-42].

This series of exothermic combustion reactions could increase the combustion temperature and the average operating pressure inside the rocket motor. The operating pressure and flame temperature are of the most important factors affecting combustion mechanism [13]. The increase in operating pressure inside the combustion chamber causes a decrease in the induction zone; consequently the flame becomes more adjacent to the burning surface as demonstrated in Figure 4.

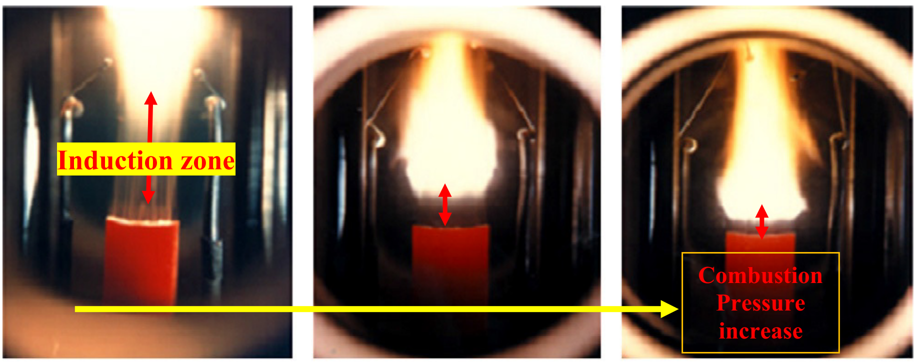


**Figure 4.** Flame behavior of double base propellant test specimen under different pressure values [43]

Aluminum particles with excellent thermal conductivity could enhance the heat transfer from the burning zone to the adjacent layers of un reacted propellant and hence decrease the thickness of the induction zone and make the luminous flame zone adjacent to the burning surface [25, 43]. The great impact of HMX on $C^*$ was attributed to HMX positive heat of formation (+353.8 kJ/kg) [31]. HMX also has only a slightly negative oxygen balance which means decomposition products of low molecular weight (M). Thus HMX can enhance the specific impulse by offering high combustion temperature ($T_c$) and low average molecular weight of gaseous products [31]. The specific impulse of solid propellant is directly proportional to the square root of combustion temperature, and inversely proportional to the square root of combustion gasses molecular weight as described by Equation 4 [25].

$$I_{sp} \sim \sqrt{\frac{T_c}{W}} \tag{4}$$

where $T_c$ is the combustion temperature and *M* is the average molecular weight of gaseous products.

## 3.3 Thermal properties of reference DB propellant formulation and developed MDB propellant formulations

Calorific value (heat of combustion) using bomb calorimeter, auto-ignition temperature using cook off test, and thermal behavior using DSC of reference DB propellant formulation and MDB propellant formulations were precisely measured and evaluated. The calorific value of the developed MDB propellant formulations was evaluated using bomb calorimeter to quantify the impact of different energetic fillers on heat released upon combustion (kJ/g). The employed bomb calorimeter consisted of two main parts; parr-6200 calorimeter and parr-6510 water handling system. 400 mg of the tested sample was completely burned up in excess oxygen (20 bar). The increase in water bath temperature (ΔT) was recorded to retrieve the generated heat upon combustion.

Auto ignition temperature is one of the main important characteristics to be evaluated [44]. The employed equipment for ignition temperature measurement was Reichel & Partner (Germany). A sample of 0.1 g was introduced in a glass tube and heated at controlled rate of 5 °C/min till ignition occurs. DSC measurements offer quantitative information about physical and chemical changes (*i.e.*, onset and maximum decomposition temperature, phase change, exothermic/endothermic decompositions). MDB propellant formulations were investigated using DSC Q-2000 (Thermo-scientific, USA). Each tested sample was heated from 50 °C to 300 °C at a heating rate 5 °C/min under $N_2$ flow at 10 mL/min. The aim of DSC measurements is to evaluate the onset decomposition temperature, total heat released (heat flow, J/g), and maximum decomposition temperature. MDB propellant formulations exhibited an increase in total heat released in comparison with the reference DB propellant formulation. The onset decomposition temperature was similar for reference DB propellant formulation and developed MDB propellant formulations. The maximum peak temperature was around 190 °C for all formulations as represented in Figure 5.

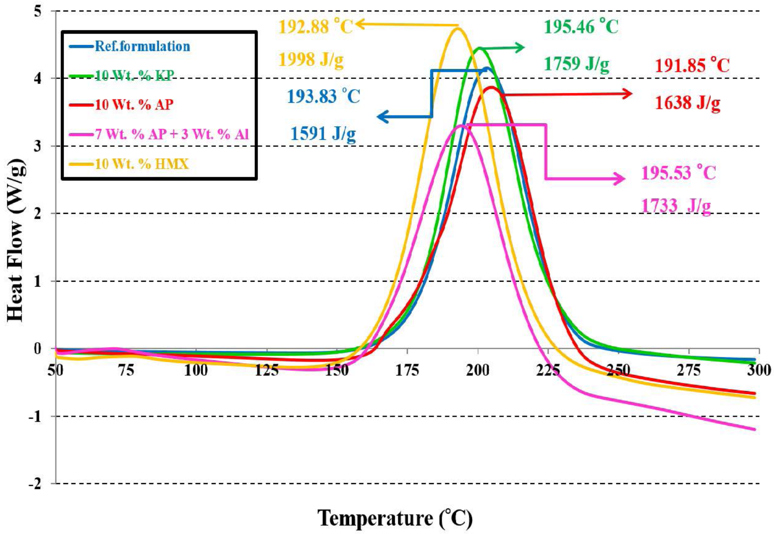


**Figure 5.** DSC thermograms of tested reference DB propellant formulation and developed MDB propellant formulations

Even though, the developed MDB propellant formulations exhibited an increase in heat released upon decomposition; they exhibited thermal behavior similar to reference DB formulation using DSC (Figure 5).

While, MDB propellant formulations offered an increase in heat of combustion (kJ/g) using bomb calorimeter; they exhibited ignition temperature and onset/maximum decomposition temperature similar to DB reference propellant formulation using cook off test and DSC measurements. For nitric ester propellants (NC and NG), thermal degradation could take place by homolytic breakdown of the O–$NO_2$ bond [45]. The inclusion of different energetic additives into DB propellant matrix did not affect its thermal degradation profile.

The main thermal properties including calorific value, auto ignition temperature, and DSC outcomes of developed MDB propellant formulations together with such properties of reference DB propellant formulation, are tabulated in Table 3.

While, MDB propellant formulations ($F_1$, $F_2$, $F_3$ and $F_4$) were found to be more energetic in comparison to the reference DB propellant formulation ($F_0$), they exhibited similar thermal behavior to the reference formulation. Formulation 4 ($F_4$) based on 10 wt.% HMX exhibited the highest heat of combustion. This could be ascribed to the fact that HMX is one of the most energetic explosive materials which could burn with the release of 30.96 kJ/g.

**Table 3.** DSC results, calorific value, ignition temperature (and density) of MDB propellant formulations and reference DB propellant formulation

| | DSC results | | Calorific value [kJ/g] | Ignition temperature [°C] | Density [g/cm$^3$] |
|---|---|---|---|---|---|
| | Total heat released [J/g] | Max. peak temperature [°C] | | | |
| $F_0$ (Reference) | 1591 | 193.83 | 3891 | 171 | 1.612 |
| $F_1$ (10 wt.% KP) | 1759 | 195.46 | 4192 | 171 | 1.625 |
| $F_2$ (10 wt.% AP) | 1638 | 191.85 | 3962 | 169 | 1.617 |
| $F_3$ (7 wt.% AP + 3 wt.% Al) | 1733 | 195.53 | 4071 | 172 | 1.619 |
| $F_4$ (10 wt % HMX) | 1998 | 192.88 | 4339 | 170 | 1.613 |

## 4 Summary and Conclusions

The impact of different energetic additives, *i.e.* potential oxidizers (KP & AP), stoichiometric binary mixture oxidizer/metal fuel, and energetic nitramine (HMX) on MDB propellants ballistic performance, was evaluated using small-scale ballistic evaluation test motor. Burning rate, characteristic exhaust velocity, and average operating pressure are the main parameters that were retrieved from small-scale test motor. The two inorganic oxidizers KP and AP exhibited different effect. Whereas, KP positively impact the burning rate by 22% increase; AP increased the $C^*$ by 5.3%. This phenomenon was correlated to the fact that the main determining step in burning process is the endothermic decomposition of inorganic oxidizer; KP decomposition is more exothermic compared to AP and it has higher oxidizing power. On the other hand, AP decomposition is accompanied with larger volume of gaseous products; thus it positively impacted the $C^*$ by 5.3%. Stoichiometric binary mixture of AP/Al enhanced both burning rate and $C^*$ by 12% and 10% respectively; this was correlated to the impact of Al as energetic fuel which would increase the thermal conductivity of the propellant as well as its heat of combustion. HMX was found to be ideal energetic filler which upon decomposition generates large amount of heat (6.197 kJ/g) and evolves low molecular weight gases. This is why HMX increased $C^*$ by 9.5%; the burning rate was almost unchanged.

On the basis of thermal experiments, MDB propellant formulations were found to be more energetic than reference DB propellant formulation but in the mean time they exhibited similar thermal behavior to the reference formulation. This paper highlights the fact that MDB propellant formulations with controlled combustion and tailored ballistic parameters were developed.

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