# Effect of Fuel Content and Particle Size Distribution of Oxidiser on Ignition of Metal-Based Pyrotechnic Compositions

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#### ABSTRACT

Influence of boron content in boron-based pyrotechnic composition and particle size distribution of oxidiser, i.e.,  $KNO_3$  in Mg-based pyrotechnic composition is examined by subjecting these to various tests. Study on boron-based pyrotechnic compositions reveals that compositions with 20, 25 and 30 parts by weight of boron are promising igniter compositions wrt their calorimetric values, pressure maximum, ignition delay, etc. However, from sensitivity point of view, the composition with 30 parts of boron is more safe to handle, manufacture and use. From the study of particle size distribution of  $KNO_3$  in Mg-based pyrotechnic compositions, it is observed that the composition with wider particle size distribution of oxidiser gives better packing density for their binary mix with metal fuel, which in turn gives lower ignition delay and ignition temperature.

## 1. INTRODUCTION

For the pyrotechnic compositions, the ignition temperature, ignition delay, pressure-time profile (PTP), calorimetric values (cal-val), and sensitivity to impact and friction are of prime importance. For pyrotechnic composition, being a solid-solid heterogeneous system, the packing of particles plays an important role. The packing density of particles influences rheological, hydrostatic, hydrodynamic, mass and energy transport, sonic, electric and optical parameters. It is sufficient to know about the bulk properties of the packed bed to assess the average properties<sup>1</sup>. Though the packing is achieved through compaction, the process can be made easier and less dangerous if the optimum range of particle size of the reactants in right proportion are present in dry mix. The optimum range of particle size is generally achieved by trial and error method by mixing differently the ground particles of the reactants in a bimodal,

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trimodal or multimodal mode.

For evaluation of these parameters, two sets of compositions, having varied amounts of fuel (boron) content and the particle size distribution of the oxidiser  $(KNO_3)$  were prepared. Boron was selected as a fuel because of its low atomic weight, high heat output, ready ignitibility with  $KNO_3$  releasing large amount of heat<sup>2</sup>, and persistence burning even at low pressure<sup>3</sup>.

#### 2. EXPERIMENTAL SETUP

#### 2.1 Details of Ingredients

Ingredients	Specifications		
B (amorphous) powder (Gr. I)	CS:5507		
Mg powder (Gr. V)	CS:5035		
KNO <sub>3</sub> (Gr. I)	IS:301-1983.		
Ethyl cellulose (type N 200)	CS:2724		
Diethyl phthalate	IND/ME/873		
Toluene (nitrating grade)	IS:537–1977		

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## 2.2 Preparation of Pyrotechnic Compositions

Requisite proportions of boron/Mg and dried KNO, of required particle size (63-90 µm for set I compositions, and 53-600, 53-300, 53-150, and 53-90 µm for set II compositions) and required quantity of plasticised ethyl cellulose (PEC) were mixed in a planetary mixer to obtain homogeneous dough. The dough was then granulated by passing through 600 um sieve and retaining on 300 um sieve. In set I. 10 compositions were prepared varying the boron content from 5 to 50 parts in 100 parts by weight, of its binary mix with KNO, while 10 parts by weight of PEC was added over it. In set II, four Mg-based pyrotechnic compositions were prepared by varying ranges of particle size distribution of KNO, having 53-600, 53-300, 53-150, and 53-90 µm size particles, keeping the percentage composition the same as 42 per cent Mg (fuel), 50 per cent KNO, and 8 per cent PEC as binder. The details of compositions are given in Table 1.

 
 Table 1. Data on Mg-based pyrotechnic compositions (Mg/ KNO<sub>3</sub>/ PEC: 42/50/8)

Comp. No.	Oxidiser particle size	densities		Heats of combus- tion	Ignition Temp. by DTA
	distri- bution (µm)	Oxidiser (g/cc)	Oxidiser & fuel mix (g/cc)	(cal/g)	(°C)
1	53-600	1.1538	1.2510	1784	572
2	53-300	1.1364	1.2310	1793	602
3	53-150	1.1029	1.1120	1800	603
4	53-90	0.8929	0.9300	1774	616

# 3. CHARACTERISATION OF COMPOSITIONS

All compositions from sets I and II were characterised for cal-val by Julius Peters' bomb calorimeter, ignition temperature by indigenously fabricated micro differential thermal analyser (DTA), friction sensitivity by Julius Peters' friction apparatus and impact sensitivity by standard fall and hammer method<sup>4</sup> to obtain height of 50 per cent explosions, ignition delay and pressure maximum by closed vessel firings<sup>5</sup> and packing density for different ranges of particle sizes of oxidiser and their binary mix with fuel by tapping method.

# 4. RESULTS & DISCUSSION

## 4.1 Calorimetric Values

Table 2 shows that cal-val increases with increase in boron content and reaches a maximum of 1884 cal/g for 20 parts of boron composition. This is in agreement with the theoretical value, as well as the results reported by Lindslay<sup>6</sup>. The cal-val decrease with further increase in boron content.

Table 2. Heat of combustion and ignition temperature data

Comp. No.	Composition mass by parts (B: KNO <sub>3</sub> )	Heats of combustion (cal/g)	Ignition temp. by DTA (°C)	
1	5:95	DNI	DNI	
2	10:90	1470	563	
3	15:85	1776	562	
4	20:80	1884	562	
5	25:75	1714	523	
6	30:70	1701	502	
7	35:65	1616	533	
8	40:60	1580	532	
9	45:55	1220	514	
10	50:50	690	510	

(DNI: Did not ignite)

For all the compositions of set II, cal-val are of the same order (Table 1). This is expected since the ratio of fuel and oxidiser is the same for all the compositions.

## 4.2 Sensitivity to Friction and Impact

All the compositions in set I are insensitive to friction up to 36 kg of load, but more sensitive to impact as compared to composition exploding (CE). Out of these, composition having 30 parts of boron is the least sensitive to impact as seen from Table 3.

Comp. No.	Composition by parts B: KNO <sub>3</sub>	Impact test height of 50% explosions (cm)	Friction tes insensitive up to (kg)	
1	5:95	82	36	
2	10:90	80	36	
3	15:85	79	36	
4	20:80	78	36	
5	25:75	80	36	
6	30:70	85	36	
7	35:65	73	36	
8	40:60	75	36	
9	45:55	75	36	
10	50:50	72	36	
11	CE	95	36	

Table 3. Sensitivity data of pyrotechnic compositionsat temperature: 29 °C and relative humidity:55 per cent

Table 4 of set II shows that composition with wider particle size distribution of oxidiser is more sensitive to impact. The sensitivity decreases while going from wider to narrower range of particle size.

## 4.3 Ignition Temperature by DTA

The results of DTA in Table 2 indicate that the composition having 30 parts of boron has the lowest ignition temperature. However, with either increase or decrease in boron content, the higher ignition temperatures are observed.

Table 4. Closed vessel firing and sensitivity data at temperature: 30 °C and relative humidity: 55 per cent

Comp. No.	Oxidiser particle size distribution range (µm)	Pressure maximum (kg/cm <sup>2</sup> )	Ignition delay (ms)	Impact test height of explosion (cm)	Fricton test insen- sitive up to load (kg)
1	53-600	46.4	65	81	36
2	53-300	50.3	95	83	36
3	53-150	50.3	193	85	36
4	53-90	51.2	235	90	36
5	CE	-	-	95	36

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For the Mg-based compositions of set II, the results of DTA in Table 1 show that the ignition temperature is the lowest (572 °C) for the composition having a wider distribution of particle size of the oxidiser. Higher ignition temperatures are observed for narrower distribution range of particle size of the oxidiser.

#### **4.4 Closed Vessel Firings**

The results of closed vessel firing in Table 5 show that the ignition delay and time to reach pressure maximum decreases progressively with increase in boron content up to 30 parts, and remains almost constant with further increase in boron<sup>3</sup>.

Table 5.	Closed	vessel	firing	results
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Boron by parts in Comp.	Pressure maximum P <sub>max</sub> (kg/cm <sup>2</sup> )	Ignition delay (ms)	Burn time to reach co P <sub>max</sub> (ms)	Mass onsumption rate (g/s)
5	25.80	630	575	-
10	47.88	125	50	140
15	51.66	50	22	318
20	52.99	25	18	388
25	47.29	23	16	437
30	42.85	20	14	500
35	38.65	21	17	411
40	34.33	21	21	333
45	31.79	19	22	318
50	28.00	22	22	318

The pressure maximum first increases with increase in boron content and reaches a maximum of 52.99 kg/cm<sup>2</sup> for 20 parts of boron composition. However, with further increase in boron content the pressure of maximum value again decreases.

The results in Table 4 for Mg-based composition indicate that ignition delay progressively increases from wider to narrower range of particle size distribution of oxidiser, i.e., 65 ms for particle size distribution 53-600  $\mu$ m to nearly four times (235 ms) for 53-90  $\mu$ m. The pressure maximum indicates that it is almost the same for all formulations.

#### 4.5 Packing Density

The wider range of particle size (53-600 µm)

distribution of oxidiser alone and its binary mix with Mg give higher packing densities, whereas lower values are observed for the narrower range. The details of packing densities and particle size distributions and spans are given in Tables 1 and 6, respectively.

The low values of ignition delay and ignition temperature contrary to high value of median height of 50 per cent explosions for set I compositions can be explained on the basis of hot spot theory<sup>4,7</sup> for impact sensitivity and thermal response of pyrotechnic composition. The ignition delay, ignition temperature and impact sensitivity are the functions of ratio and intimate contact of fuel and oxidiser, their particle sizes, packing densities, heat capacities, exothermacities and heat flow by conduction within the pressed mix.

For set II compositions, the low values of ignition delay and ignition temperature for wider particle size distribution of oxidiser can be explained on the basis of geometrical modelling and packing densities. In çase of wide distribution range of particle size of oxidiser, the packing density of composition is higher as the oxidiser with varieties of particle sizes are available to fill up the interstitial cavities, and thereby making the oxidiser and fuel particles in intimate contact with each other, and hence decreasing the ignition temperature and ignition delay. However, the packing density of composition with narrow range of particle size distribution of oxidiser is comparatively lower as the interstitial cavities between oxidiser-oxidiser, fuel-fuel and oxidiser-fuel might be left open or getting filled with the inert binder in the ternary mix, in the absence of a variety of particle sizes; thereby making oxidiser and metal-fuel contact less intimate, which results in increase in the ignition delay and the ignition temperature. The packing problem is well attempted by Hermance<sup>8</sup> by considering the curvilinear triangle formed by three large circles.

## 5. CONCLUSION

For an efficient igniter, the cal-val, pressure maximum and mass consumption rate should be reasonably high, whereas the ignition delay, ignition temperature and time to reach pressure maximum should be minimum. Over and above, the composition fulfilling these requirements should also be processable and safe to handle.

From the results of set I, though the compositions with 20, 25 and 30 parts of boron seem to be attractive igniter compositions, the composition with 30 parts of boron is superior considering safety aspects and its better pelleting properties.

From the observations for Mg-based compositions, it can be concluded that for faster initiation of an igniter composition, the fuel and oxidiser particles should be in intimate contact with each other. Wider the particle size distribution of an oxidiser, more

Composition	Particle size (µm)			Span	Average	
No.	53-90	90-150	150-300	300-600		(µm)
1	54.1 % {90 % <307	23.9 % μm, 10 % <41 μ	12.2 % m}	9.8 %	2.97	127
2	59.3 % {90 % < 18	26.5% 0 μm, 10 % < 41	14.2% μm}	-	1.59	106
3		37.6 % μm, 10 %<41 μm	}	-	1.12	90
4	100 % {90 % < 12	1 μm, 10 % < 30	- 7 μm}	-	1.26	75
Mg	100 % {90 % < 88	- .7 μm, 10 % < 23	- 3.4 μm}	-	1.26	55

Table 6. Analysis of particle size distribution of KNO, and Mg powder by Malvern particle size analyser

is the packing density of its binary mixture with Mg. This leads to the reduction in ignition temperature and ignition delay of the igniter. Also from the particle size distribution study by Malvern analyser, it is observed that when the spans of particle size distribution are of the same order for both fuel and oxidiser, then the packing density is lower, resulting in higher ignition delay and ignition temperature.

# REFERENCES

- Haughey, D.P. & Severidge, G.S.G. Structural properties of packed beds – A review. Can. J. Chem. Engg., 1972, 47, 130-40.
- Conkling, John A. Chemistry of pyrotechnics: Basic principles and theory. Marcel Dekker Inc., New York, 1985. 72 p.
- Barnes, P.; Griffith, T.T.; Charsley, E.L.; Hider, J.A. & Warrington, S.B. The properties and reaction of *B/KNO*<sub>3</sub> pyrotechnic systems. Proceedings of Eleventh International Pyrotechnic Seminar, 1986, Vail Colorado. IIT Research Institute Chicago, Illinois. p. 27.
- 4. Services text book of explosives JSP-333. Chapter 14, Para 42.Controller of Her Britannica Majesty's Stationary Office, UK, 1972.

- Dillehay, D.R. Closed/bomb testing at Longhorn ammunition plant. Proceedings of the Fifth International Pyrotechnic Seminar, 1976, Denver, Colorado. National Technical Information Services (NTIS), Springfield, VA-22161. p.107-22.
- Lindselay, G.I.; Robinson, E.A.; Charsley, E.L. & Warington, S.B. A comparison of the ignition characteristics of selected metal /oxidant systems. Proceedings of the Eleventh International Pyrotechnic Seminar, 1986, Vail Colorado. IIT Research Institute Chicago, Illinois. pp. 425.
- Bonnett, D.L. & Butler, P.B. Hot spot ignition of condensed phase energetic materials. J. Propu. Power, 1996, 12 (4), 680-90.
- 8. Hermance, C.E. On the dense packing of spherical particles, views of packing space and introductory discussion of particle size distribution. Proceedings of the Sixteenth International Pyrotechnic Seminar, 1991, Jonkoping, Sweden. Sektionen for Detonik. Och Forbranning Sundbyberg, Sweden. pp. 347-58.



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