

## Nitramine-Based High Energy Propellant Compositions for Tank Guns

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### ABSTRACT

Six different RDX-based gun propellant compositions have been formulated and studied to select the most suitable composition for tank gun ammunition in terms of higher force constant at relatively lower flame temperature ( $T_f$ ). Ballistic performance of the compositions was evaluated on the basis of closed vessel test. Heat energy was determined using a bomb calorimeter. Sensitivity, thermal characteristics, stability and mechanical properties of the compositions were studied for assessing their suitability for application. The composition containing 65 per cent RDX and 28 per cent nitrocellulose was found to provide higher level of force constant at relatively lower  $T_f$ , reasonably good burning rate characteristics and mechanical properties.

### 1. INTRODUCTION

Conventional gun propellants have reached saturation level in terms of energy. To meet the requirements of tank gun ammunition, propellants of higher force constant at a relatively lower flame temperature ( $T_f$ ) are required to minimise gun barrel erosion. Linear rate of burning coefficient  $\beta$ , and pressure exponent ( $\alpha$ ) are important parameters in determining the combustion behaviour and suitability of a propellant for tank gun ammunition. A higher value of  $\beta$ , necessitates increase in web size of the propellant grain. Increase in web size, in turn, poses problems in manufacture, loadability and brittle fracture of the grain, particularly at lower temperature. Similarly, a higher magnitude of  $\alpha$  leads to exponential rise in burning rate and pressure, which affect the safety of the gun.

Extensive research is being carried out all over the world to improve upon the force constant by increasing the number of moles of combustion gases per unit mass rather than increasing  $T_f$ . The main constituents of the propellant combustion gases are carbon monoxide, carbon dioxide, hydrogen,

water and nitrogen. To formulate a higher energy gun propellant, the ingredients should have higher percentages of hydrogen, carbon monoxide and nitrogen rather than water and carbon dioxide in their combustion product gases. Literature survey reveals that a number of new series of cool burning, high impetus and low molecular weight gun propellants have been studied<sup>1-4</sup>. Most propellants contain either cyclic or linear nitramines, such as cyclotrimethylene-trinitramine (RDX), cyclotetramethylene-tetranitramine (HMX), triaminoguanidine nitrate (TAGN), nitroaminoguanidine (NAGU) and triaminoguanidine-ethylene-dinitramine (TAGED) as energetic ingredients. The results of a systematic study on RDX-based propellant compositions aimed at obtaining higher energy for tank gun applications are presented in this paper.

### 2. EXPERIMENTAL WORK

Six different compositions based on nitrocellulose (NC) of 13.1 per cent nitrogen content, di-octylphthalate (DOP), carbamate and RDX of average particle size (5  $\mu\text{m}$ ) were formulated. Theoretical performance of the composition was computed using THERM

**Table 1. Chemical formulations and theoretical performance of propellant compositions**

Parameter	Composition number					
	II	III	IV	V	VI	
NC (13.1N%)	36	32	28	24	20	16
DOP	8	7	6	5	4	
Carbamite						
RDX (5 $\mu$ m)	55	60	65	70	75	80
Flame Temp (K)	2942	3075	3210	3342	3469	3585
Force constant (J/g)	1132	1168	1202	1235	1265	129
$P_{max}$ (MPa)	283	291	299	307	313	319
Co-Volume (ml/g)	1.0040	0.9939	0.9837	0.9734	0.9631	0.9523
'n' Value (moles/g)	0.04628	0.04567	0.04505	0.04445	0.04386	0.04331
Sp. heat ratio (r)	.2645	1.2662	1.2599	1.2577	1.2558	1.2542

program and the results are presented<sup>5</sup> in Table 1. Propellant compositions were made on a laboratory scale (1 kg batch) by solven process<sup>6</sup>. First, fine RDX was dehydrated with ethyl alcohol and coated with required quantity of DOP on dry weight basis. Exact percentage of DOP and uniformity of its coating to RDX was confirmed gravimetrically using *n*-pentane as the solvent for the extraction of DOP. All the samples were subjected to impact and friction sensitivity tests to obtain safety-related information. Propellant compositions were prepared using a 30 per cent solution of acetone-alcohol (70:30) mixture. The ingredients were kneaded in an incorporator for 6 hr to obtain a homogeneous propellant dough. Five per cent extra solvent was required to have a good dough during the preparation of compositions containing 75-80 per cent RDX. The dough was subsequently extruded in cord form at around 50 bar using a hydraulic press. Extruded cord strands were cut to 12 cm and dried in an oven at 45-50 °C till the volatile matter got reduced to 1 per cent. Out of the six compositions, composition III, containing 65 per cent RDX and 28 per cent NC, was made in a multitubular configuration also

**Table 2. Data on closed vessel firing of the propellant compositions**

Parameter	Composition number						
	II <sup>+</sup>	III <sup>+</sup>	IV <sup>+</sup>	V <sup>+</sup>	VI <sup>+</sup>	VII <sup>+</sup>	
Web size (mm)	1.65	1.66	.67	1.70	1.71	1.73	1.50
Propellant density (g/cc)	.60	.61	.62	1.62	1.63	1.65	1.60
$P_c$ (MPa)	280	290	297	312	323	330	
Force constant (J/g)	1130	1165	1200	1244	1272	1305	1192
Linear burning rate coefficient $\beta_1$ (cm/s/MPa)	0.12	0.13	0.14	0.24	0.30	1.11	0.12
Pressure exponent ( $\alpha$ )	1.07	1.09	1.10	1.41	1.44	2.12	0.84

+ Cord configuration

\* Multitubular configuration

Loading density = 0.20 g/ml; propellant temperature = 27 °C

to study its burning rate characteristics in that configuration. Dried propellant samples were tested for physical characteristics like web size and density, and finally fired in a 700 cc closed vessel (CV) at 0.20 g/cc loading density for the determination of ballistic performance. Results of CV tests are shown in Table 2.

**Table 3. Data on sensitivity of the propellant compositions**

Sensitivity test	Composition number					
	II	III	IV	V	VI	
Impact (height for 50 % explosion) (cm)	50.0	46.0	44.5	43.5	43.0	35.0
Friction (insensitive up to) (kg)	28.8	25.2	25.2	24.0	24.0	21.6

Reference sample = Composition exploding (tetryl)

Relative humidity = 55-60 %

Room temperature = 29 °C

### 3. EVALUATION OF BALLISTIC PERFORMANCE

#### 3.1 Measurement of Sensitivity

Impact sensitivity was measured by fall hammer method using 2 kg drop weight and 20 mg sample. The height mentioned in Table 3 refers to 50 per cent probability of explosion of the compositions. Friction sensitivity was measured using Julius Peter apparatus and 10 mg sample. The results for impact and friction sensitivity are given in Table 3.

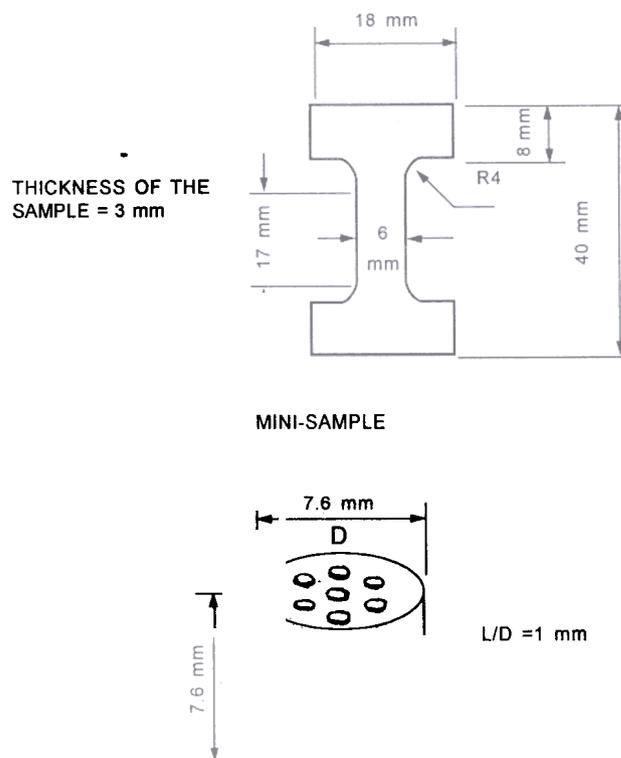
#### 3.2 Thermal Characteristics

Deflagration temperature was obtained on 5 mg sample by gradually raising the temperature at the rate of 5 °C/min in Julius Peters furnace. The temperature at which the sample got ignited was recorded. Decomposition temperature was recorded using differential thermal analyser (DTA). DTA curves were recorded in an inert atmosphere using 10 mg samples in alumina crucibles at a heating rate of 10 °C/min. Calorimetric values of the compositions were determined using Julius Peters adiabatic bomb calorimeter of 300 cc at 1 atm. 1g sample was ignited and the total heat output was measured (Table 4).

**Table 4. Data on thermal characteristic tests of the propellant compositions**

Test	Composition number					
	II	III	IV	V	VI	
Ignition* temperature (°C)	200	> 200	> 200	200	200	200
DTA decomposition temperature and its range (°C)	194 164-206	196 168-206	199 174-210	201 185-215	222 173-240	222 178-240
Nature of DTA exotherm (sharp)	One exotherm (sharp)	One exotherm (sharp)	One exotherm (sharp)	One exotherm (sharp)	Two exotherm (one sharp & one weak)	Two exotherm (one sharp & one weak)
Calorimetric value (cal/g)	904	923	987	1035	1084	1138

\* In case of ignition temperature test, all the six compositions did not ignite even up to 350 °C. Around 200 °C, samples started producing yellow fumes.



**Figure 1. Specimen samples for testing the mechanical properties.**

#### 3.3 Thermal Stability

Thermal Stabilities of the compositions were determined by Bergmann and Junk test. 5g samples were heated at 120 °C for 5 hr and the total gaseous volume of nitrogen oxides was measured titrimetrically.

#### 3.4 Mechanical Properties

For determining tensile strength and percentage elongation, 'mini-samples' (Fig. 1) were punched out of the propellant strips and dried up to ~1 per cent V.M. level. Tensile strength and percentage elongation were determined using the Instron universal materials testing machine (model-1185). Flexural properties were determined using propellant strips of particular dimensions (100 mm × 10 mm × 3 mm) and Instron machine. For determining percentage compression of the composition containing 65 per cent RDX and 28 per cent NC, multitubular grains having  $L/D = 1$  (Fig. 1) were made. The Instron machine was used for this purpose.

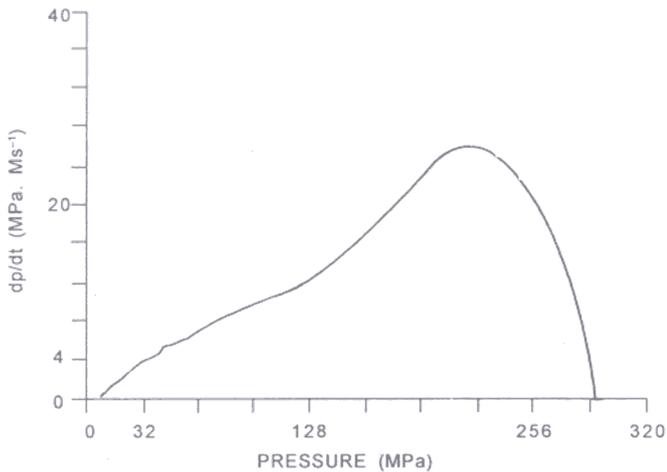


Figure 2. P vs  $dp/dt$  profile of composition III (multitubular configuration).

#### 4. RESULTS & DISCUSSION

The results of ballistic evaluation of the propellant compositions using a  $cv$  are given in Table 2. Compositions I and II exhibited comparatively lower values of force constant. However, composition

III was found to be most suitable because of its higher force constant at a relatively lower  $T_f$ .  $\beta_1$  value was found to be within acceptable limit, but  $\alpha$  value exceeded unity. Therefore, the same propellant composition was further studied in a multitubular configuration to assess its suitability wrt  $\beta_1$  and  $\alpha$ . Results of  $cv$  tests on the subject composition indicate 14 per cent and 23 per cent decrease in the value of  $\beta_1$  and  $\alpha$ , respectively when the cord configuration was changed to multitubular format. This difference appears to be due to approximation in the form function for multitubular shape and also ignition and burning characteristics of holes of the multitubular propellant<sup>7</sup>. Approximation in the form function is achieved due to anomalous combustion mechanism of RDX propellant, as indicated by the slope break phenomenon. Slope breaks are related to the change in the mechanism of decomposition, probably due to changes in depth of melt-layer of the deflagrating propellant surface. The form function is thus suitably approximated in the multitubular rather than the cord configuration, which results in a low pressure exponent. Therefore, suitability

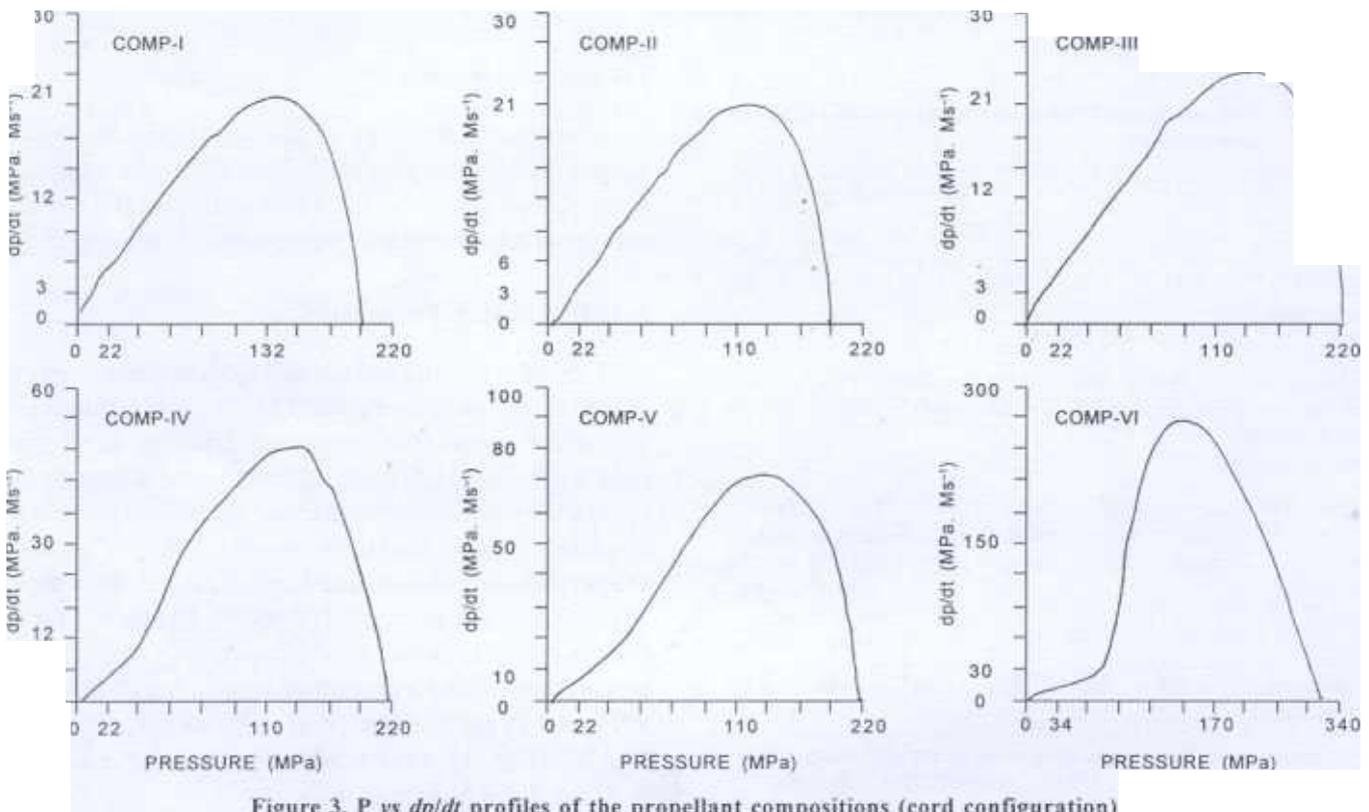


Figure 3. P vs  $dp/dt$  profiles of the propellant compositions (cord configuration)

of the subject composition for tank gun applications was better assessed when it was manufactured in a multitubular configuration. Secondly,  $P_{vs}$   $dp/dt$  profile of the multitubular propellant (Fig. 2) was found to be of non-peaky nature as compared to the cord configuration. For compositions containing 70, 75 and 80 per cent RDX, even though the force constant increased successively, values of  $T_p$ ,  $\beta_1$ , and  $\alpha$  also increased drastically for beyond the desired levels for tank gun applications. Hence, these compositions have not been selected for tank gun applications. The same is confirmed by  $cv$  firing, wherein a sudden rise in  $P_{vs}$   $dp/dt$  profile was noticed in the case of composition containing 80 per cent RDX and 16 per cent NC (Fig. 3).

The measured sensitivity values given in Table 3 indicate increasing trend of sensitivity from the compositions containing 55 per cent RDX and 36 per cent NC – 80 per cent RDX and 16 per cent NC. This may be attributed to a successively increasing order of oxygen balance<sup>8,9</sup>.

Data on deflagration temperature (Table 4) indicate that gradual decomposition of NC extended from 180 °C to 200 °C, as yellow vapourisation was observed around 200 °C. DTA curves of the first four compositions containing 55 per cent RDX and 36 per cent NC – 70 per cent RDX and 24 per cent NC show uniformity and single mass decomposition, as only one exotherm was recorded. The regular increase in the decomposition temperature is attributable to successively increased solid loading of RDX within the NC matrix. For the last two compositions containing 75 per cent RDX and 20 per cent NC and 80 per cent RDX and 16 per cent NC, respectively, two exotherms have been recorded. The first exotherm at 203 °C for the former and at 207 °C for the latter indicate decomposition of propellant mass formed with the maximum loadable RDX within the NC matrix, whereas the second exotherm at 222 °C for both the compositions indicates decomposition of surplus RDX, which could not be loadable within 20 per cent and 16 per cent of NC matrix, respectively (Fig. 4). This observation indicates nonsuitability of these two compositions for tank gun applications.

Calorimetric value was found to increase regularly, indicating a successively higher energy output due

Table 5. Data on thermal stability (Bergmann and Jank test at 120 °C) of the propellant compositions

Observation	Composition number					
	I	II	III	IV	V	VI
Quantity of gas evolved (ml/5 g)	0.3	0.25	0.25	0.25	0.20	0.10

Specification: Quantity of gas evolved > 5 ml/5g

to the more exothermic reaction between NC and increased content of RDX in comparison to the compositions containing 55 per cent RDX and 36 per cent NC – 80 per cent RDX and 16 per cent NC (Table 4).

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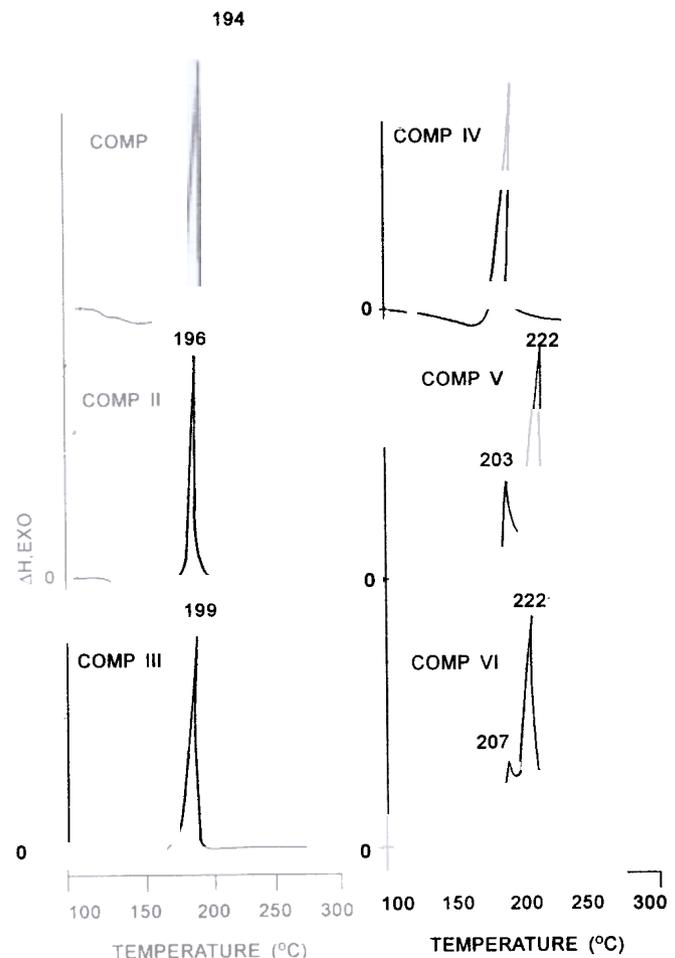


Figure 4. DTA curves

of the subject composition for tank gun applications was better assessed when it was manufactured in a multitubular configuration. Secondly,  $P_{vs} dp/dt$  profile of the multitubular propellant (Fig. 2) was found to be of non-peaky nature as compared to the cord configuration. For compositions containing 70, 75 and 80 per cent RDX, even though the force constant increased successively, values of  $T_p$ ,  $\beta_1$ , and  $\alpha$  also increased drastically for beyond the desired levels for tank gun applications. Hence, these compositions have not been selected for tank gun applications. The same is confirmed by *cv* firing, wherein a sudden rise in  $P_{vs} dp/dt$  profile was noticed in the case of composition containing 80 per cent RDX and 16 per cent NC (Fig. 3).

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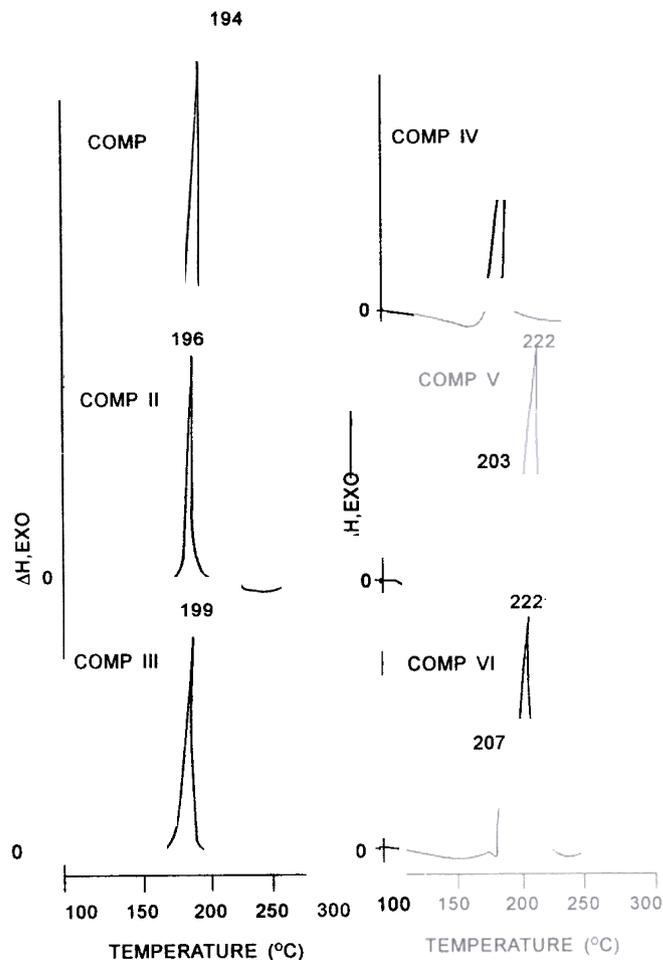


Figure 4. DTA curves

**Table 6. Data on mechanical properties of the propellant compositions**

Mechanical property	Composition number					
	II	III	IV	V	VI	R
Tensile strength (kg/cm <sup>2</sup> )	226	217	180	95	Sample got broken at the grips while fixing	Sample got broken at the grips while fixing
Percentage elongation	5.14	5.60	3.66	3.05	·do·	·do·
Flexural test (displacement at yield) (mm)	4.26	2.83	.53	.27	1.06	0.85
Compression tests (percent strain at maximum load)	–		13.10			10.33

R = Standard NQ propellant

was higher in comparison to compositions containing 55 per cent RDX and 36 per cent NC – 80 per cent RDX and 16 per cent NC. This may be attributed to decreasing gaseous volume of nitrogen oxides, as the NC content decreased successively.

Data on mechanical properties given in Table 6 indicate the tensile strength and percentage elongation to be in the decreasing order from the composition containing 55 per cent RDX and 36 per cent NC – 70 per cent RDX and 24 per cent NC. However, mini-samples of the compositions containing 75 per cent RDX and 20 per cent NC – 80 per cent RDX and 16 per cent NC got broken at the grips, while fixing the jaws of the Instron machine during the course of experiment. This is indicative of poor mechanical properties. Additionally, the flexural test indicates decreasing order of flexibility in terms of displacement at yield from the composition containing 55 per cent RDX and 36 per cent NC – 80 per cent RDX and 16 per cent NC. This may be attributed to successively lower contents of NC and DOP, which are responsible for mechanical properties. The availability of long carbon-carbon chains in the molecular structures of NC and DOP causes absorption of strains and stresses, contributing towards

improved mechanical properties. Higher value of percentage compression for the selected composition containing 65 per cent RDX and 28 per cent in NC in comparison to the conventional NQ propellant indicates potentiality of its use in tank guns, particularly at high solid loading.

## 5. CONCLUSION

The propellant composition III containing NC (13.1 per cent nitrogen) DOP/carbamite/RDX (5  $\mu$ m) has been found to provide higher force constant (1200 j/g) with relatively lower  $T_f$  (3210 K), reasonably good burning rate characteristics and mechanical properties.

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### Contributors



**Mr RS Damse** obtained his MSc (Organic Chemistry) from University of Poona, in 1983. He joined DRDO at the High Energy Materials Research Laboratory (HEMRL) as Scientist B. His areas of work include: development of high explosives for filling of various warheads, design and development of combustible cartridge cases for tank guns of different calibre and ignition system for tank gun ammunition.



**Dr Hardiwar Singh** obtained his PhD in solid rocket propellant combustion from University of Poona. He joined DRDO at the HEMRL, Pune, in 1971 and became Director in 1990. He is a recognised post-graduate guide of University of Poona and Shivaji University and has supervised 20 post-graduate and doctoral theses. He was a Visiting Scientist at the Max-Planck Institute, Germany; High Pressure Combustion Laboratory, Pennsylvania State University, Army Research Laboratory; Maryland Science Application Centre, Santa Clara and Sandia National Laboratory, USA. He has been awarded the *Aeronautical Society of India Award* for 1994. He was conferred with the *DRDO Scientist of the Year Award* in 1983 for his contributions in applied sciences and advanced solid propellants, high energy propellants, respectively. He is the recipient of *DRDO Cash Award* for the development of power plants for missile target and *Best Paper Award of Defence Science Journal* in 1984. He is the Chairman of the High Energy Materials Society of India, a member of AIAA, International Advisory Board-Flame Structure & Combustion, and an honorary member of the Russian Academy of Astronautics. His areas of research include: rocketry, cast double-base (CDB) propellants, HTPB-based composite propellants, nitramine-based propellants, high energy composite modified double base (CMDDB) for integrated rocket ramjet, AP-based propellants, and zirconium powder based fuel-rich propellants.