Effects of Explosive Crystal Internal Defects on Projectile Impact Initiation

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Summary

Based on accurate apparent density measurements, experiments have been performed to correlate the explosive crystal microstructure and the sensitivity of cast formulation. The experiments are based on commercial grade RDX and on ISL RDX crystal lots with very few internal defects. Cast formulations containing 70 weight percent of RDX and 30 percent of wax as a binder have been processed and impacted with a 20 mm projectile.

As with HMX in a previous paper, a strong correlation is recorded between the formulation sensitivity and the volume fraction of intra-crystalline voids. The critical kinetic energy to get the detonation increases from 21 kJ to 35 kJ when the intragranular void volume reduces from 0.44% to 0.06%.

The residual extragranular void volume of the formulation is accurately studied. No clear trend with the formulation sensitivity is observed, despite some variations (0.06% to 2%). The effects of extragranular voids and particle sizes are lower than the effects of intragranular voids. The role of pore sizes is discussed. The coarse ISL RDX crystals, with very few intragranular pores, have a sensitivity closer to the sensitivity of ultrafine RDX (5 µm) than all other RDX lots.

INTRODUCTION

The importance of effects of microstructure on the shock sensitivity of explosive compositions has been well known for many years[1]. Heterogeneities in the explosive material lead to the formation of hot spots, where the first chemical reaction occurs. A shock sensitivity control of the explosive formulations needs to identify the hot spots and to understand their mechanisms[2] .

Numerous studies over the years have shown that the presence of porosity in an explosive formulation sensitizes it to shock loading. Pores can occur in the binder material, at the binder-explosive crystal interface, and within the explosive crystals themselves.

A lot of studies have been devoted to investigate the effects of explosive crystal characteristics on the shock-detonation transition of cast plastic bonded explosives. The explosive crystal properties studied were: the size [3], the shape and the surface [4 5], the defects at different scales inside the explosive crystals [6 7]. Despite all these works, it is not yet possible to describe accurately the separate role of these various microstructure features. The main experimental difficulties are the lack of measurement tools for an accurate characterization of the microstructure of the explosive composition and the lack of processing control to manufacture customized explosive particle lots.

Using the ISL sink-float method and RDX defect free crystals processed in ISL [8], this paper provides new quantitative and accurate experimental data. The ISL sink-float method is a unique, accurate, and quantitative tool to measure the apparent densities of the explosive particles and to characterize the intragranular void volume of an explosive particle lot [9].

This paper is organized in four parts. The first part provides additional data for previous HMX based experimental data. The second part introduces the RDX crystal lots used in this study. The cast formulations employed are detailed and accurately characterized. The third part gives the RDX shock sensitivity experimental results. The last part is a discussion pointing out the new results.

PREVIOUS RESULTS BASED ON HMX CAST FORMULATIONS

Using cast formulations containing 70 weight-percent HMX (200-300 microns) and wax as binder, it was reported [6] that the projectile velocity threshold for detonation decreased by 30 percent as the total intra-crystalline void volume increased from 0.1 percent to 0.45 percent. As no quantitative data were reported for the formulation density, an ambiguity remained in regard to the amount of the extragranular porosity [2].

Table 1 provides new quantitative data for the amount of extragranular voids in the cast HMX formulations. The total intra-crystalline void volume for the explosive crystal lot is accurately computed from the apparent density distribution measured using the ISL sink-float method. This provides also the true apparent density of the explosive lot.

HMX (200-300 µm) 70% + wax 30%	Lot 1	Lot 2	Lot 3
Projectile impact velocity threshold (m/s)	1100	830	730
Projectile impact Kinetic energy threshold (kJ)	30.2	17.2	13.3
Intragranular void volume (% / unit crystal volume)	0.15	0.3	0.45
Extragranular void volume (% / unit formulation volume)	0.3	0.6	0.001

Table 1: HMX experimental results

The use of wax as a binder, with no chemical reactions during the formulation processing allows the accurate computation of the theoretical maximum density of the formulation knowing the wax density and the true apparent density of the explosive lot. The total extragranular void volume is then computed from the measured formulation density and the computed theoretical maximum density.

The new data of table 1 confirm the strong correlation between the intragranular porosity and the shock sensitivity of the formulations. The values of the extragranular void volume fraction do not correlate simply with the formulation shock sensitivity but they are coherent with the qualitative observations of the shape of the HMX crystals. Lot 3 exhibits particles with more spherical shapes and particles of lot 3 are easier to cast that particles of lot 1 and lot 2. Particles of lot 3 provide the best formulation, free of extragranular voids (0.001%) but particles of lot 3 lead to the most sensitive formulation. This shows that the remaining extragranular voids have a minor effect on the formulation sensitivity.

In order to check these first results, we have performed the same kind of accurate experiments on similar RDX based formulations.

Three features of the RDX crystal microstructure are studied: the crystal sizes, the crystal surface properties and the crystal internal defects.

RDX CRYSTAL LOTS AND CAST FORMULATIONS

RDX crystal lots

Two kinds of RDX crystal lots have been used :

- RDX lots processed in the ISL laboratory. The main interest of the ISL RDX lots is a full control of the crystal properties to test accurately the separate effect of the various crystal properties on the formulation sensitivity.
- Commercial grade RDX. Their use allows to widen the range of variation of the RDX crystal properties because the processing methods employed can be very different. The counterpart is a lack of control of some crystal properties as the processing methods employed are not precisely known.

RDX lot	Particle sizes (µm)	Intragranular pore volume (%)
Lot 1	100/315	0.20
Lot 2	100/315	0.53
Lot 3	100/315	0.44
Lot 4	100/315	0.12
Lot 5	100/315	0.24
Lot 6	100/200	0.14
Lot 7	100/200	0.14
Lot 8	315/800	0.10

Table 2:commercial grade RDX

Table 2 gives the crystal properties of the commercial grade RDX. Some lots have been sieved from commercial grade RDX to reduce the particle size range in order to work with the same narrow monomodal particle size distribution.

The lots 1 to 5 have different crystal surface properties (crystal shape, surface porosity). Accurate measurement tools[5] are missing for a direct and accurate characterization of the crystal surface properties. The accurate control of the cast formulations provides an indirect measurement of the crystal surface properties. This will be detailed further.

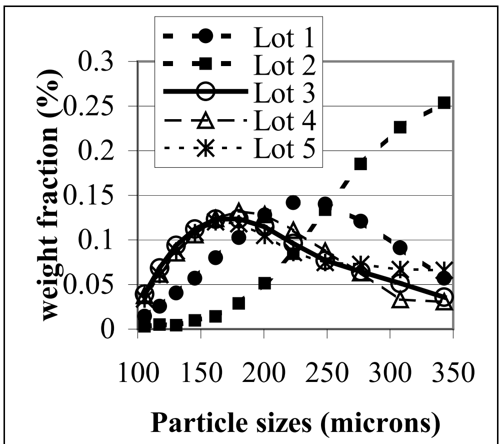
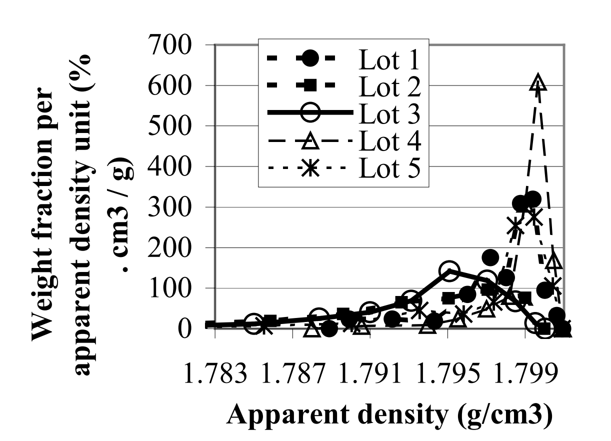


Figure 1 : Particle sizes of commercial grade RDX

The particle size range of lots 1 to 5 is 100-315 microns (figure 1). The sizes repartitions are similar excepted for lot 2.

These five lots have different crystal internal defect populations, which have been accurately characterized using the ISL float-sink method (figure 2).Table 2 provides the volume fraction of pores inside the crystals. This fraction refers to a unit volume of the RDX crystals. Lot 4 have the most narrow apparent density distribution and lots 2 and 3 have the widest apparent density distributions among lots 1 to 5.



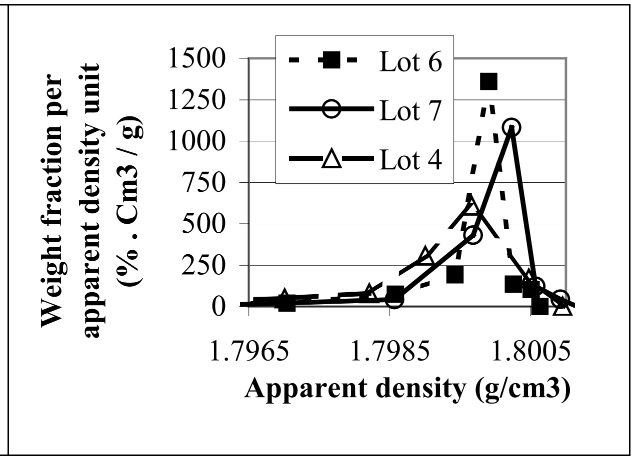


Figure 2 : Apparent density distribution of commercial grade RDX

Figure 3: Apparent densities distributions of lots 6 and 7.

Lots 6 and 7 have similar apparent density distributions (figure 3). Lots 6 and 7 have the same narrow and monomodal particle size range:100-200 µm. Crystals of lot 7 has been processed from crystals of lot 6 in order to get smoother surfaces and more spherical shapes. Lots 6 and 7 have different crystal surface properties (figure 4).

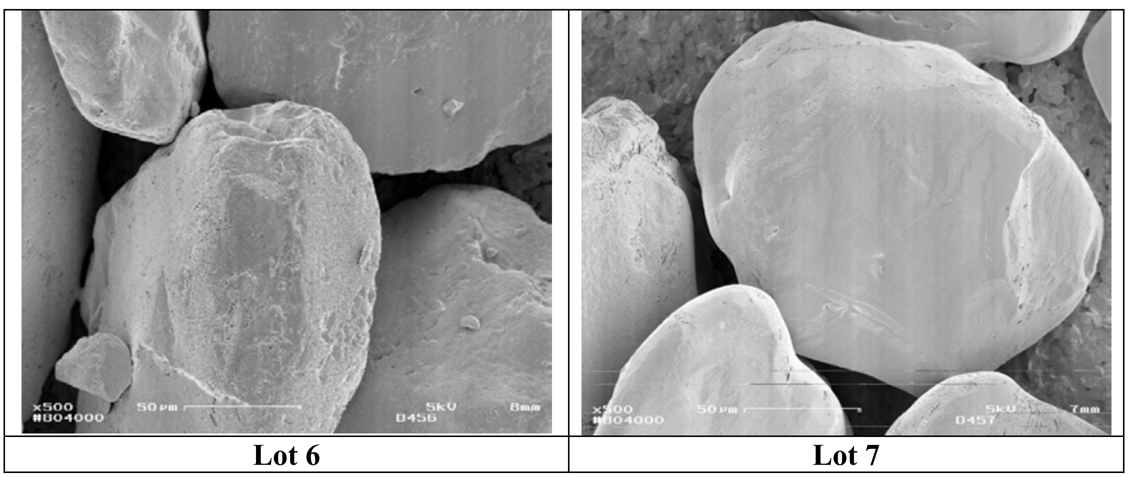


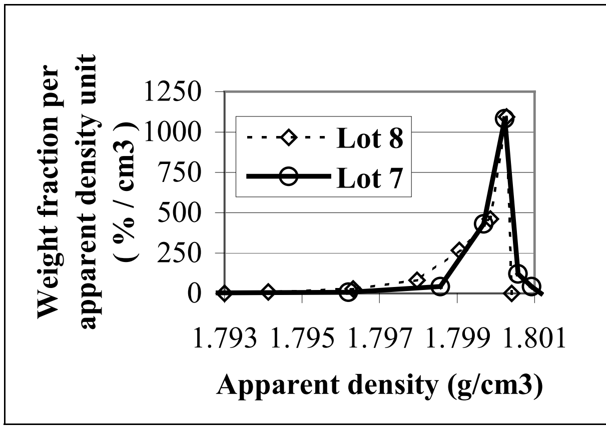
Figure 4: Crystal surface properties of lot 6 and lot 7.

The particle size range of lot 8 is 315/800 microns. This lot has been selected because of a very low amount of internal defects (figure 5). Lot 7 and Lot 8 have similar apparent density distribution curves but not the same particle sizes.

RDX lot	Particle sizes (µm)	Volume fraction of pores inside the crystals (%)
ISL 1	100 / 315	0.06
ISL 2	315 / 630	0.06
ISL 3	100 / 630	0.06

Table 3 : ISL laboratory RDX lots

A same process have been used for ISL RDX lots. The use of different sieves provides three ISL lots , with three different particle size ranges (table 3). These lots have been crystallized in order to get a very low amount of internal defects.



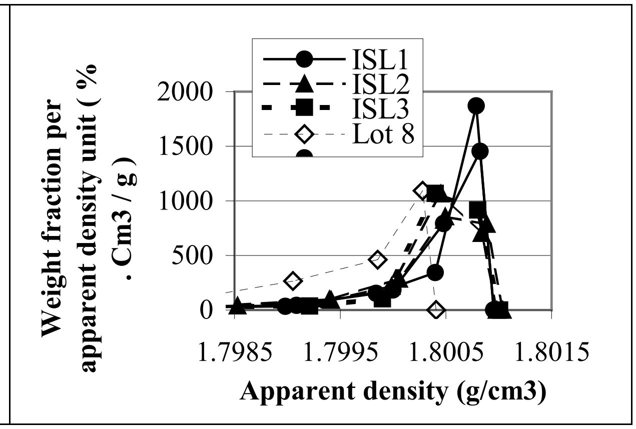
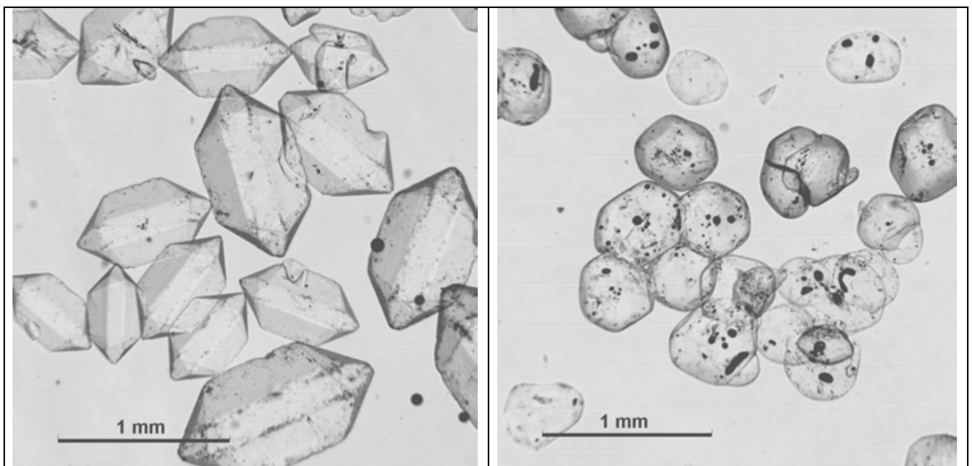


Figure 5 : Apparent density distribution of lot 8.

Figure 6 : Apparent densities of ISL lots



ISL2 (315/630 microns)	Lot 8 (315/800 microns)

Figure 7 : optical microscopy with matching refractive index

The very low amount of internal defects of the ISL lots (figure 7) is confirmed quantitatively by the crystal apparent density distribution measurements (figure 6). The crystal apparent density distributions are monomodal, with a very narrow and high mode (figure 6). These ISL crystal lots have the best properties. Crystal surface properties of the ISL crystals are different from the eight previous RDX lots. The surface porosity is reduced. The crystal shapes are those of single crystals with characteristic flat faces and marked edges (figure 7).

Cast formulations

All the shock sensitivity tests have been performed using similar cast formulations. The weight solid load is 70 percent. A same wax is used as inert binder. Only the explosive crystal lot varies between the formulations. Similar formulations have been used in previous experimental studies [5,6,9,10].

The formulation is processed by casting under gravity and the cooling is performed under controlled conditions. This provides homogeneous raw cylinders, from which the targets for the sensitivity tests are extracted. The use of explosive crystal lots with narrow monomodal particle size distribution and very different particle surface properties leads to some variations of the residual amount of extragranular voids despite the use of 30% in weight of binder.

The use of wax, a non-polymeric binder, allows an accurate and quantitative checking of the formulation density. The density of the raw cast cylinder is recorded by weighting the cylinder in air and immersed in water. The amount of voids external to the explosive crystals is determined by comparison of the measured apparent density and the theoretical maximum density (TMD). The theoretical maximum density of the formulation is computed using the apparent density of the explosive particle lot deduced from the measured apparent density distribution. The volume fraction of extragranular voids refers to a unit volume of the cast formulation (table 4).

RDX lot	**Volume fraction of extragranular voids (%)**
Lot 1	0.38
Lot 2	0.06
Lot 3	0.80
Lot 4	1.95
Lot 5	0.36
Lot 6	2.29
Lot 7	1.11
Lot 8	0.23
ISL1	1.63
ISL2	0.8
ISL3	1.22

Table 4 : cast formulations

The formulations using the ISL RDX lots were among the most difficult to cast. The flat surface and marked edges of the ISL RDX crystals can provide an explanation for that behavior.

With ISL RDX lots, the amount of residual extragranular voids increases when the particle size is reduced from 315-630 µm (ISL 2) to 100-315 (ISL1). This shows that the amount of residual voids increases with the specific surface area. This trend is confirmed with ISL3 whose particle size range is 100-630 µm and which exhibits an intermediate amount of extragranular voids.

The effects of crystal surface properties is confirmed also with lots 6 and 7. Lots 6 and 7 have the most narrow particle size range 100/200 µm and the smallest average particle size. Lot 6 leads to the highest volume fraction of extragranular voids. Lot 7 is obtained from lot 6 using a crystal surface processing to get more spherical and smooth crystals. This leads to reduce by a factor 2 the volume fraction of extragranular voids. It is coherent to use the volume fraction of extragranular voids as an indirect measurement of the crystal surface properties of an RDX lot.

SMALL PROJECTILE IMPACT EXPERIMENTAL RESULTS

The reactive behavior of the cast formulations is tested by impacting formulations samples with a flat ended steel projectile of diameter 20 mm, length 20 mm and mass 50 g. The formulation sample diameter is 42 mm and its length is 50 mm. The shock transit time across the sample is recorded for several projectile impact velocity. The sharp decrease of the shock transit time, when the sample detonates, provides the impact velocity threshold to get a full detonation of the sample. More details on the experiments are available in [6].

Sustained plane shock wave have shown that the effects of the explosive crystal microstructure were recorded only for the lowest shock pressures [10]. This can be understood using the hot spot concept. At low shock pressures, the size threshold needed for a cavity to become an efficient hot spot is higher than at high shock pressure. This limits the number of potential hot spots and points out the explosive crystal microstructure effects.

The small projectile impact experiment is a low shock pressure experiment [6]. In addition, the small projectile impact introduces lateral effects which reduce the number of ignited sites and increase the effects of the explosive crystal microstructure [6, 9].

For each RDX lot (each formulation), 8 formulation samples have been used to check the reproducibility of the experiments and to determine the projectile impact velocity threshold. Table 5 provides the velocity and the kinetic energy thresholds of the impacting projectile to get the detonation of the formulation sample for the various RDX lots used.

Lot	**Velocity threshold (m/s)**	**Kinetic energy threshold (kJ)**
Lot 1	1096	30
Lot 2	1073	29
Lot 3	919	21
Lot 4	1067	28
Lot 5	1061	28
Lot 6	1044	27
Lot 7	1124	32
Lot 8	1180	35
ISL1	1177	35
ISL2	1187	35
ISL3	1176	35

Table 5: small projectile impact: experimental results.

The kinetic energy threshold of the projectile exhibits large variations (from 21 to 35 kJ) in function of the RDX lot employed. As for HMX [6], this underlines the important role of the RDX crystal microstructure on the shock sensitivity of cast high explosive formulations.

Figure 8 plots the projectile kinetic energy threshold in function of the volume fraction of intragranular voids (table 2 and 3). Reducing the amount of intragranular voids (lots 8, ISL1, ISL2, ISL3), increases the projectile kinetic energy threshold to get the detonation of the formulation. This means that the formulation shock sensitivity increases when the volume fraction of intragranular voids increases. The ISL RDX lots show that the different crystal sizes (100-315 and 315-630) have no effect on the shock formulation sensitivity.

Figure 9 plots the projectile kinetic energy threshold to get the detonation in function of the volume fraction of extragranular voids. No clear trend is pointed out for the projectile kinetic energy threshold in function of the amount of extragranular voids.

Both RDX and HMX experimental results exhibit the same trend : a dominant role for the intragranular voids. A more detailed analysis of this trend is performed in the following part.

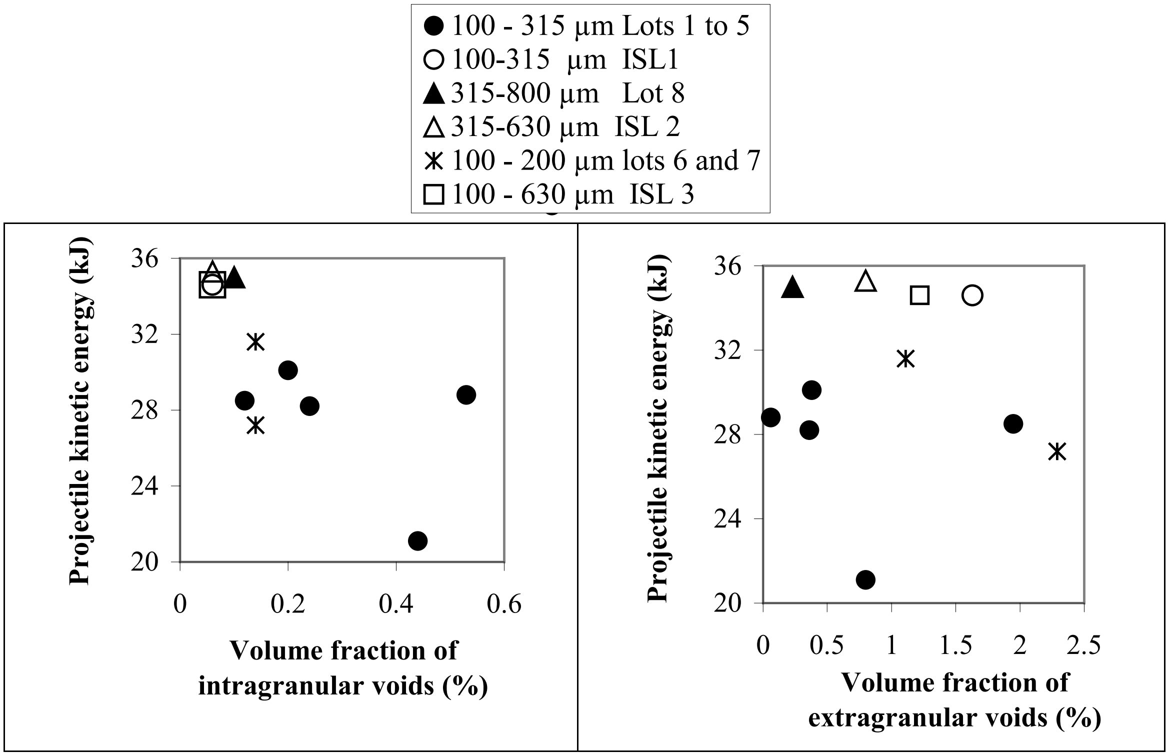


Figure 8 : effects of intragranular voids

Figure 9 : effects of extragranular voids

DISCUSSION

Figure 8 points out a more clear trend (intragranular voids) than figure 9 (extragranular voids). Lot 3 has one of the highest volume fraction of intragranular voids and gives the most sensitive formulation (21 kJ). Lot 8 and ISL lots (ISL1, ISL2, ISL3) have the lowest level of intragranular voids and give the lowest level of formulation sensitivity (35 kJ).

Lots 1, 4, 5, 6, 7 have about the same volume fraction of intragranular voids, similar crystal sizes and about the same intermediate projectile kinetic energy threshold (30 kJ), but these lots have very different volume fraction of extragranular voids. RDX crystals of lot 7 are RDX crystals of lot 6 which have been processed in order to smooth the crystal surface and get more spherical crystals. Details on this surface processing are not known, but this surface processing leads to divide by 2 the volume fraction of extragranular voids. This leads to an increase of the kinetic energy threshold from 27 kJ to 32 kJ, but the main trend remains. An increase of the volume of intragranular voids gives an increase of the formulation sensitivity.

Nevertheless, some deviations are recorded with commercial grade RDX (lots 1 to 8). This needs further investigations as for instance for lot 2. It exhibits the largest volume fraction of intragranular voids and an intermediate projectile kinetic energy threshold (29 kJ). Measurements of the pore sizes (for both intragranular and extragranular porosities) could provide explanations. Lot 8 shows also a deviation. Apparent densities of crystals of lot 8 are lower than apparent densities of ISL crystals (figure 6), but the same kinetic energy thresholds are recorded.

A possible explanation is an effect of extragranular voids. Lot 8 has a smaller volume fraction of extragranular voids than ISL lots. The numerical computations performed by Conley and co-authors [11] point out the important role of the intragranular voids in cast PBX free of extragranular voids. They show a similar role of extragranular voids and intragranular voids when the binder is removed. The shock to detonation transition is the result of two competitive processes : local heating by hot spots, and thermal loss by conduction. Even if the temperature of extragranular hot spots is too low and lower than the temperature of intragranular hot spots, the heating of the extragranular hot spots can reduce the thermal losses by conduction and contribute to enhance the shock detonation transition. It would be interesting to perform an extension of the computations to our case: a PBX with intragranular and extragranular voids.

With commercial RDX lots, a more detailed analysis of the experiments is difficult. The manufacturing processes are not known. A unique and known manufacturing process is used for the ISL lots and the dominant role of intragranular voids is confirmed. ISL lots exhibit the same volume fraction of intragranular voids and the same projectile kinetic energy threshold. But ISL lots have different particle size distributions and different volume fraction of extragranular voids. With ISL RDX lots extragranular voids have no effects on the projectile kinetic energy threshold.

Assuming that the sizes of the extragranular voids remains the same as the casting process used is always the same, ISL RDX lots show that the sizes of the extragranular voids are small. These sizes are lower than the critical size for a pore to become an efficient hot spot at a given shock pressure.

It is very interesting to compare these new RDX data with those published by Moulard and co-authors [12]. The same sensitivity experiment (20 mm steel projectile) were performed on similar cast formulations : 30% polyurethane as a binder + 70 % (weight) RDX. Two RDX lots had been used. A coarse RDX lot with a particle size range 100-200 µm and a fine RDX lot with a particle size range 0-12 µm. The projectile velocity threshold to get the detonation for the formulation using the coarse RDX lot was about 1090 m/s (30 kJ). This is the sensitivity recorded with our formulations using lots 6 and 7 (100-200 µm). This allows to compare our new data with the low sensitivity recorded by Moulard for the fine RDX (5 µm) lot. The projectile velocity threshold to get the detonation for the formulation using the fine RDX lot was about 1295 m/s (42 kJ). Our new experimental data show that using coarse particle sizes (100-315 µm or 315-630 µm) with a lower amount of intragranular voids than commercial grade RDX , the usual low pressure sensitivity level can be improved from 30 kJ to 35 kJ. This is an intermediate level between the fine RDX used by Moulard and usual coarse RDX.

CONCLUSION

Our experimental data points out the role of explosive crystal intragranular voids. The data recorded with ISL RDX lots confirm and improve the trend recorded with commercial grade RDX and HMX. Removing most of the intragranular pores (ISL lots) provides cast formulations with a sensitivity level which does not depend on crystal size, nor on the extragranular voids of the formulation. This sensitivity level is closer to the sensitivity level of ultrafine RDX (5 µm) than all other RDX lots.

These first results needs to be confirmed and improved. The ISL attempt to reduce intragranular voids in RDX crystals is promising for both academic studies and applications.

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