## CRITICAL DIAMETER OF LIQUID EXPLOSIVES AS A FUNCTION OF POWDER CONTENT

## R. Kh. Kurbangalina

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The dependence of the critical diameter (d\*) of nitromethane (NM) on the content of aluminum, aluminum oxide, tungsten powders, carbon black, and talc and the dependence of the d\* of tetranitromethane (TNM) on aluminum and aluminum oxide content have been experimentally investigated. The powder content was varied over a wide range (0-75% by weight), as was the particle size. It was found that for NM mixtures the variation of d\* is quite different from that for TNM. For powder particle sizes of  $1-50 \mu$  the d\* of the NM mixtures decreases with increase in powder concentration. The minimum value of d\* is ten times less than the value for pure NM. In TNM mixtures d\* increases monotonically with the amount of powder. It is assumed that this behavior of the NM mixtures is associated with the inhomogeneous structure of the detonation front in NM, a consequence of the particular reaction kinetics characteristic of nitromethane.

The critical diameters were determined in the usual way in long glass tubes with walls 0.6-1.3 mm thick. The temperatures of the rooms in which the charges were prepared and tested were the same (+20° C) with deviations of not more than +2 and -1° C. The detonation process was registered on film by means of a high-speed photorecorder and also by the deformation of a metal plate positioned beneath the charge. To prevent settling of the powder particles, the solutions of TNM and NM were first thickened.

Certain data on the properties of the TNM, NM, the thickened starting solutions, and the powders employed are presented in the Tables 1 and 2, where the density, heat of explosion at constant volume, and the critical diameter are denoted by  $\rho$ ,  $Q_v$ , and  $d_*$ , respectively.

Table 1				
Explosive	₽, g/cm³	$Q_v$ , kcal/kg	<i>d</i> <sub>*</sub> , mm	
Nitromethane NM + colloxylin 97/3 Tetranitromethane TNM + plexiglas 99.5/0.5	1.139 1.14 1.638 1.63	1060—1472 542 574	$\begin{array}{c c} 12.5\\ 12\\ 10.8-11\\ 9.6 \end{array}$	

The experimental results on the  $d_*$  of the mixtures are presented in Figs. 1-3, where along the axis of abscissas we have plotted the powder content in percent by weight (P), and along the ordinate axis the charge diameter (d) in mm. In all cases a plus sign denotes the propagation of detonation, and a minus sign the absence or quenching of detonation. The value of  $d_*$  for a mixture of given composition is located between the nearest minus and plus for symbols of the same kind.

Table 2	
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Powder	p, g/cm3	Characteristic
Al	2.7	Particle size ( 0.2 $\mu$
Al	2.7	ASD-1; particle size basically 5-10 $\mu$
W Al <sub>2</sub> O <sub>3</sub> Al <sub>2</sub> O <sub>3</sub> Al <sub>2</sub> O <sub>3</sub> Al <sub>2</sub> O <sub>3</sub>	$ \begin{array}{r}     19.3 \\     3.9-4.1 \\     3.9-4.1 \\                                    $	basically 1 $\mu$ Corundum MP-7, 7 $\mu$ Corundum MP-1, 1 $\mu$ Anal. gr. TUMKhT2063-49, $\rangle$ 50 $\mu$ For chromatography, $\rangle$ 50 $\mu$
Carbon black	1.8 - 2.1	Fine
Talc	2.7 - 2.8	1-100 μ

The data for the mixture TNM + Al ASD-1 are presented in Fig. 1 (points 1). The critical diameter of the TNM only increases with increase in the aluminum content of the mixture. This result is somewhat unexpected: there are no extrema, although an extremum of  $d_*$  might be anticipated when the heat release of the mixture (stoichiometric composition) is at a maximum. This suggests that the aluminum in the TNM behaves with respect to  $d_*$  as an inert powder. To confirm this we performed several experiments on TNM + Al<sub>2</sub>O<sub>3</sub> 7 $\mu$ , denoted in Fig. 1 by symbols 2. The values of

 $d_*$  for this mixture are distributed along the curve (though slightly below it), thereby confirming that the aluminum behaves like an inert powder.

In Fig. 2 there are two curves. Curve 1 represents the variation of the critical diameter of NM as a function of the content of ASD-1 aluminum, curve 2 is for the mixture  $NM + Al_2O_3 7\mu$ . Curves 1 and 2 in this figure differ completely from the curve in Fig. 1; in Fig. 2 for both mixtures  $d_*$  decreases catastrophically as the powder content increases; curves 1 and 2 are cup-shaped and, consequently, have a minimum, where  $d_*$  is 5-10 times less than the value for pure NM. This behavior may be described as anomalous. Moreover, the value of  $d_*$  for the mixture NM + + powder 25/75 always remains less than the  $d_*$  of the pure NM.



The data plotted in Fig. 3 represent the results of experiments conducted principally to determine the effect of the particle size and density of the powders on the d\* of NM + powder mixtures; curve 1 (symbols 1)-NM +  $Al_2O_3$  7  $\mu$ , curve 3 (symbols 3)-NM +  $Al_2O_3$  1  $\mu$ , curve 2 (symbols 2) NM + W 1  $\mu$ , symbols 4-NM +  $Al_2O_3 > 50 \mu$ , symbols 6-NM +  $Al < 0.2 \mu$ , symbols 5-NM + carbon black.



The most interesting property of the data in Fig. 3 is the dependence of the critical diameter of the NM + powder mixture on the particle size, which is clearly expressed in connection with the mixtures NM + Al<sub>2</sub>O<sub>3</sub> and NM + Al; the lowest curve in Fig. 3 is curve 1 for NM + Al<sub>2</sub>O<sub>3</sub> 7 $\mu$ ; the data for both NM + Al<sub>2</sub>O<sub>3</sub> 1 $\mu$  (curve 3) and for NM + Al<sub>2</sub>O<sub>3</sub> > 50 $\mu$  (symbols 4) lie much below the data for NM + Al<sub>2</sub>O<sub>3</sub> 7 $\mu$ . The value of d<sub>\*</sub> for the mixture with very fine aluminum (symbols 6 in Fig. 3) is also much higher than the value for the mixture NM + Al ASD-1 (Fig. 2, curve 1). Thus, the least values of d<sub>\*</sub> correspond to the mixtures of NM and Al<sub>2</sub>O<sub>3</sub> powder with a particle size of 7 $\mu$ . Whether precisely this particle size is the critical size ensuring minimum values of d<sub>\*</sub> cannot be confirmed without performing experiments with Al<sub>2</sub>O<sub>3</sub> powders with particle sizes differing somewhat from 7 $\mu$  in both directions. However, of all the Al<sub>2</sub>O<sub>3</sub> powders tested so far (particles sizes 1, 7, and > 50  $\mu$ ), 7- $\mu$  aluminum oxide reduces the value of d<sub>\*</sub> more sharply than all the rest.

High values of  $d_*$  (symbols 5 in Fig. 3, and an increase in  $d_*$  as the carbon black content increases are noted in connection with the mixture NM + carbon black. The chief reasons for this are apparently the excessive dispersity (<0.1  $\mu$ ) of the carbon black particles and their relatively low density.

To determine the effect of the hardness of the powder on  $d_*$  we performed special experiments on NM mixed with tale: these showed that other things being equal the  $d_*$  for NM + tale and NM + Al<sub>2</sub>O<sub>3</sub> mixtures coincide, i.e., the hardness of the powder does not have much influence on the investigated effect.

This research was carried out in order to determine the values of d\* for TNM and NM in mixtures with various powders, since an accurate estimate based on calculation is not yet possible. According to the ideas developed in [1], the quantity  $d_{\star}$ , the reaction time  $\tau$  of an explosive in the detonation zone, and the temperature T in the layer of liquid explosive compressed by the shock front are related:  $d_* \sim \tau \sim \exp A/RT$ . For single compression of the liquid by the detonation front the temperature T is determined by the velocity D of the detonation wave:  $T \sim D^2$ . When inert substances are added to the explosive, the detonation velocity is always reduced; consequently, T is also reduced. A reduction in T should lead to an increase in d\*. This is approximately the situation with mixtures of liquid explosive + inert liquid. As these experiments show, the behavior of mixtures of liquid explosive + powder with respect to  $d_*$  is anomalous. The system liquid explosive + powder is a heterogeneous medium; the laws of propagation of shock and detonation waves in such a system are more complicated than in the system liquid explosive + inert liquid, which is homogeneous with respect to the state of aggregation. In the liquid explosive + powder system the propagation of the shock and detonation waves is complicated both by the different compressibilities and sound velocities of the components and by the possibility of some sort of interaction between the powder particles and the shock front. When shock and detonation waves propagate through a liquid explosive + powder mixture, the temperature and pressure field (when the powder particles are not very small) is inhomogeneous, there are regions of elevated temperatures and pressures. If a region at the elevated temperature  $T_1$  reaches a certain size, the explosive in that region will ignite earlier and thus becomes a reaction center. Now the controlling temperature is no longer T, but  $T_i > T$ ;  $T_i$  is less easily estimated. Of course, the efficiency of the reaction centers is associated with their size and number. In [2] the effect of adding (up to 5% by weight) heavy inorganic finely-ground powders to certain condensed explosives on their critical diameter was investigated, and it was found that d\* decreases with increase in powder content. As a result of an analysis of their experimental data the authors of [2] came to the conclusion that the centers with elevated temperatures and pressures that developed during the propagation of a detonation wave in a condensed explosive + powder system are effective if the detonation reaction of the starting pure explosive proceeds according to a heterogeneous (focal) mechanism, i.e., when the detonation front in the starting explosive is inhomogeneous. The authors of [3-5] showed that the detonation front in NM is inhomogeneous: during detonation the NM ignites at individual centers, whereas the detonation front in tetranitromethane is homogeneous: the shock compressed layer of TNM ignites simultaneously at all points. The ideas developed in [2] are evidently also applicable to the case of liquid explosive + powder mixtures; combined with the results of [3-5] they help to clarify the different behavior of NM and TNM with respect to d\* in mixtures with powders. Nitromethane, which requires centers at elevated temperatures and pressures for the detonation reaction, obtains additional reaction centers when a powder is introduced.



The appearance of a large number of additional centers sharply reduces the ignition time of NM in the detonation front, which results in a decrease in  $d_*$ . It follows from the data of Fig. 3 that a reduction in the  $d_*$  of NM is observed only when the particle size of the powder lies in a certain range. This means that not all centers are effective. Perhaps a relation exists between the effective size of the center and the size of the inhomogeneities in the detonation front of NM. This question remains to be explored and may provide material for a further investigation.

In the case of TNM, on the other hand, introducing a powder disturbs the homogeneity of the detonation front, prevents the uniform propagation of detonation, and thereby changes the ignition mechanism. Apparently, the change in the ignition mechanism and the reduction in the total heat of explosion of the system as powder is added lead to an increase in the ignition time and hence the value of  $d_*$ . Admittedly, the critical diameters of mixtures of TNM and

powders have not yet been sufficiently investigated, especially mixtures with powder of varying particle size and high density. Moreover, the heat of explosion of TNM itself is low (see Table 1) as compared with NM. When the heat of explosion of the NM + powder mixture becomes less than 600 kcal/kg, the  $d_*$  of the system also begins to increase (see Figs. 2, 3), but in this case the absolute value of the  $d_*$  of the mixture is still less than the  $d_*$  of pure NM. This may cast doubt on the validity of our principal conclusion—the dependence of the value of the  $d_*$  of the liquid explosive + powder system on the structure of the detonation front of the starting liquid explosive. It appears that even after additional investigation the behavior of a TNM + powder mixture with respect to  $d_*$  will be different from the behavior of a nitromethane mixture, since in Figs. 1–3 the corresponding values of  $d_*$  vary in opposite directions.

A few words about the detonation velocities of the systems investigated. In explosive + inert powder systems, just as in mixtures containing a condensed phase in the explosion products, the detonation velocity is always lower than in the pure starting explosive, and the greater the proportion of condensed phase, the lower the velocity. NM + powder and TNM + powder systems behave in the same way with respect to detonation velocity. We determined the detonation velocity at the same time as we estimated the critical diameter. Accordingly, for most compositions we measured the detonation velocity only near the critical diameter, and these values have been omitted. However, for the systems NM +A1 ASD-1 and NM + Al<sub>2</sub>O<sub>3</sub>, 7. $\mu$  of 63/37 composition we recorded detonation velocity curves up to  $d/d_* = 10$ ; these are presented in Fig. 4, where curve 1 relates to NM + Al and curve 2 to NM +Al<sub>2</sub>O<sub>3</sub>. At the same compositions by weight the detonation velocity of the system NM + Al is somewhat higher than the detonation velocity of the system NM +Al increases slowly, but continually with increase in charge diameter, which may be attributed to partial combustion of the aluminum in the NM reaction zone.



In general, with respect to the nature of the dependence of detonation velocity on charge diameter liquid explosive + powder systems are closer to powdered explosives; it is clear from Fig. 4 that the detonation velocity  $D_*$  at the critical diameter is much lower (10-12%) than the D remote from the critical diameter. The existence of this similarity was previously noted in [6] in determining the critical initiation pressure in a NM + carborundum system of 20/80 composition. It was shown that, as in powdered explosives, when detonation of this system is initiated by a shock wave whose velocity is lower than the normal detonation velocity of the mixture, there is a gradual increase in the detonation velocity of the mixture until it reaches the normal value; the critical pressure of the initiating shock for a NM + carborundum mixture is equal to 23 kbars, whereas for the initiation of detonation in pure NM under the same conditions a shock wave with an intensity of 86-90 kbars is required.

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