

Non-ideal behaviour of ammonium nitrate based high-energetic materials in small diameter steel tube

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Abstract

The size effect curve for non-ideal behavior is the relationship between the detonation velocity and the inverse radius of the cylindrical charge. In this paper we discuss the size effect curves obtained by a steel tube test for an ammonium nitrate (AN)-based explosive. The results for AN and mixtures of AN and activated carbon (AC) are examined. The composition of AN/AC and the charge diameter are varied from 0.1 to 20 wt% and from 5.7 to 65.9 mm, respectively, and their effect on the detonation velocity is investigated using curved-front theory. The size effect curve for pure AN has a point of inflection, and the curve can be expressed as two straight lines. Finally, the size effect curves for AN/AC mixtures are constructed and formulated for almost the entire experimental region.

Keywords : detonation velocity, non-ideal detonation, steel tube test, curved-front theory, size effect

1. Introduction

There are two types of rapid energy release from high-energetic materials, i.e., ideal and non-ideal detonation. If the relaxation time to attain the thermodynamic equilibrium state is very short, the Chapman-Jouguet (C-J) hypothesis can be applied to estimate the propagation of detonation waves of high-energetic materials. In such a case, the rapid reaction is called ideal detonation. The ratio of the length of the reaction zone to the charge radius is important in determining the detonation behavior. In general, the detonation velocity depends on the size of the charge diameter. With increasing charge diameter, the velocity approaches the value for an infinite charge diameter. This effect is called the size effect and is one of the most important research topics related to detonation physics and remains an unsolved problem. Explosives that have a strong size effect are called non-ideal explosives which include industrial explosives, such as ammonium nitrate (AN)-based explosives, the subject of this study.

The pioneers in this field are Jones¹⁾ and Eyring et al.²⁾. Eyring et al. summarized the detonation velocity for

various charge sizes as a function of the inverse of the radius using curved-front theory. Since then, non-ideal behaviors have attracted many researchers of detonation physics, and numerous studies describing the non-ideality have been reported³⁾⁻²⁰⁾. For instance, the Wood and Kirkwood model³⁾ employs the relationship between the curvature at the detonation front and the detonation velocity. In recent studies, the detonation shock dynamic (DSD)⁸⁾ has been applied to non-ideal detonation phenomena^{18),20)}.

The non-ideal detonation of industrial explosives such as AN-based explosives is one of our research topics²¹⁾⁻²⁷⁾, along with studies on their high non-ideality. Understanding of the non-ideality is important from the viewpoints of engineering applications and explosive safety. Experiments on non-ideal behaviors employ a cylindrical sample with or without confinement. An unconfined sample must be large to achieve a steady state detonation. On the other hand, in the confined case, although the results exhibit both size and confinement effects, relatively small samples can be used in the

experiments. In addition, the confinement conditions may realize a wide range of reaction phenomena. In this study, steel tube tests on mixtures of AN and activated carbon (AC) were conducted to investigate their non-ideal behavior. Curved-front theory is used to examine the experimental data. The effects of the size, confinement, and composition on the detonation velocity of AN/AC mixtures are clarified. Finally, the size effect curves for AN/AC mixtures are constructed.

2. Experiment and data analysis of size effect

In the steel tube tests conducted to examine the effects of the composition, and charge diameter, and confinement, the mixing ratio of AN/AC was varied from 0.1 to 20 wt% AC. Pure AN was also used. The physical properties of AN and AC are respectively shown in Tables 1 and 2. Figure 1 shows the experimental setup for the steel tube tests. The length of the steel tube is 350 mm. Ten steel pipes of Japanese industrial standards, whose diameters and thicknesses are shown in Table 3, were used. The detonation velocity was measured with ionization probes placed at intervals of 50 mm. The booster explosive consists of 50 g composition C4 (density: $1.4 \text{ g}\cdot\text{cm}^{-3}$, detonation velocity: $7.05 \text{ km}\cdot\text{s}^{-1}$), which was used to induce a steady state detonation in the samples.

The relationship between the detonation velocity and the mixing ratio of AC obtained using a 35.5-mm-diameter steel tube is shown in Figure 2. The detonation velocity increased with increasing mixing ratio of AC, but above a mixing ratio of 5 wt% AC no significant change was observed. Therefore in this paper, the results for mixing ratios from 0.1 to 5 wt% of AC are examined. The relationship between the detonation velocity and initial density obtained using a 41.2-mm-diameter steel tube is shown in Figure 3. The scattering of the initial density is small. Even so, the ratio of the detonation velocity to that at an infinite charge diameter is used to discuss the size and confinement effects. The detonation velocity at an infinite charge diameter was estimated using the Cheetah

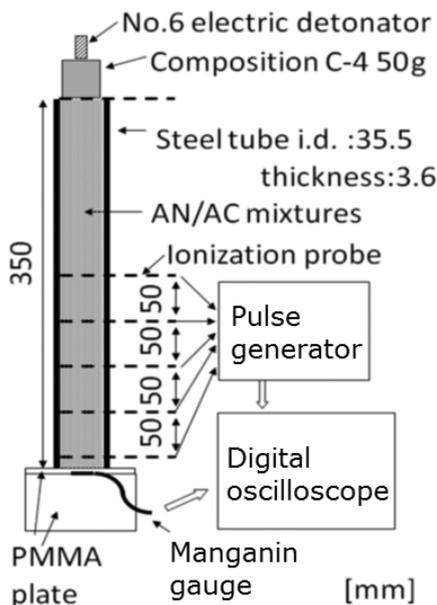


Figure 1 Schematic illustration of steel tube test.

Table 1 Physical properties of AN in this work.

AN density [$\text{kg}\cdot\text{m}^{-3}$]	Ave. dia. of particle [mm]	Purity [%]
930	0.14	> 99

Table 2 Physical properties of AC in this work.

AC density [$\text{kg}\cdot\text{m}^{-3}$]	Ave. dia. of particle [nm]	Spec. surface area [$\text{m}^2\cdot\text{g}^{-1}$]
260	3.41	1433

Table 3 Inner diameter and thickness of the steel tube; d : diameter [mm], t : thickness.

D [mm]	5.7	7.8	10.9	15.3	20.4	27.2	35.5	41.2	52.7	65.9
T [mm]	2.4	3.0	3.2	3.2	3.4	3.4	3.6	3.7	3.9	5.2

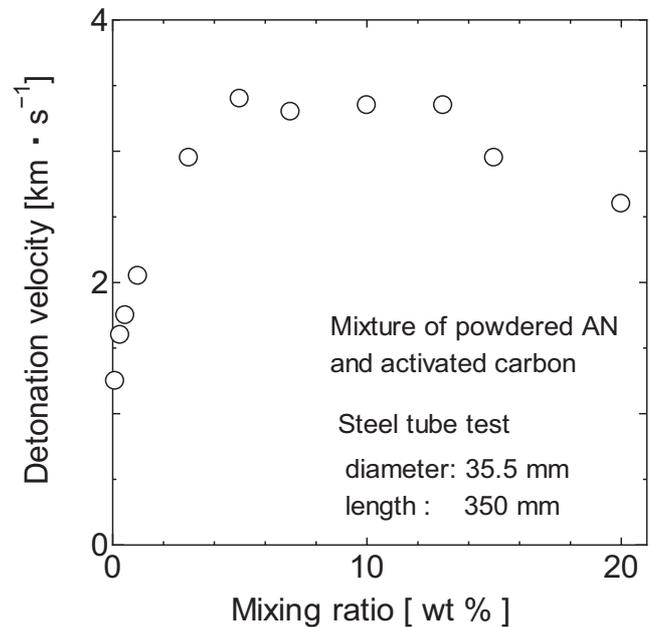


Figure 2 Detonation velocity of ammonium nitrate/activated carbon compositions vs. mixing ratio of AC obtained by steel tube test.

code²⁸⁾ for all results as shown in Figure 4.

Eyring *et al.* proposed the curved-front theory for a real detonation wave. The theory assumes that any small portion of the detonation wave can be approximated by a spherical detonation wave. The conservation law at the detonation front and generalized C-J condition were adopted to formulate the following relationship between the velocities of the real and ideal detonation waves with a reaction zone length of a .

$$\frac{D}{D_i} = 1 - k \frac{a}{R} \quad (1)$$

where, D is the velocity of the real detonation wave, D_i is that of the ideal detonation. R is the charge radius, and k is a constant. For the confined case, this relationship has been rewritten using the mass per unit areas for the explosive (We) and the confinement (Wc).

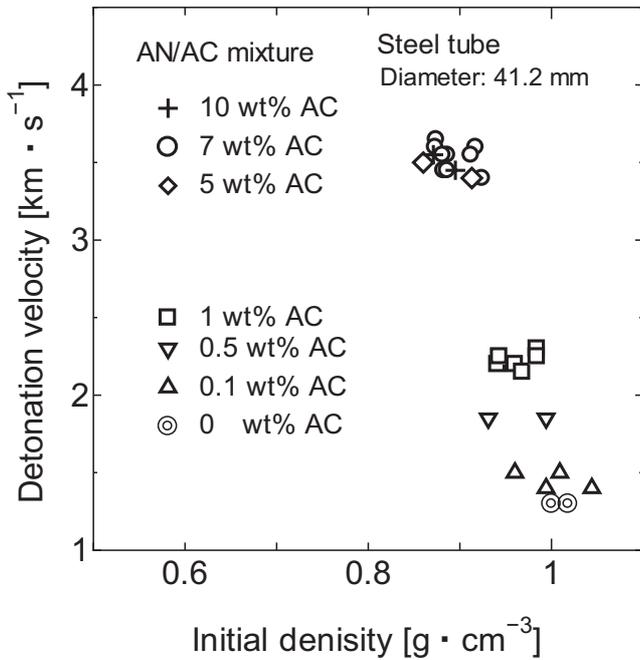


Figure 3 Detonation velocity of ammonium nitrate/activated carbon compositions vs. the initial density obtained by steel tube test.

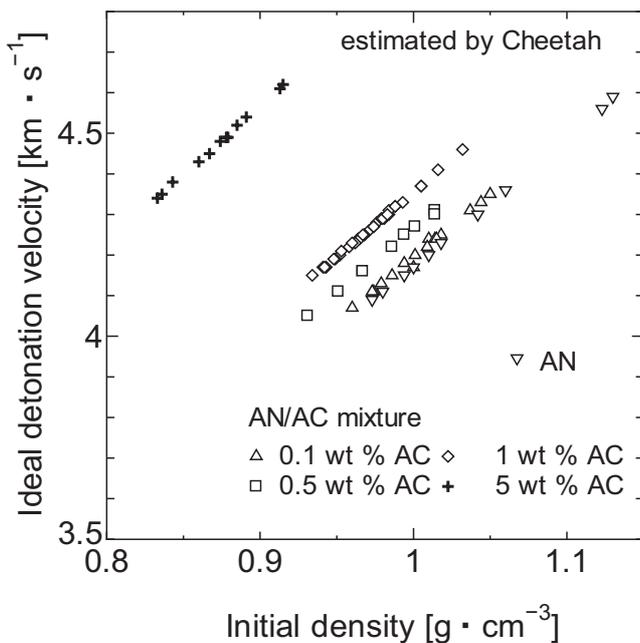


Figure 4 Detonation velocity at the infinite charge diameter estimated by Cheetah code for corresponding experimental data.

$$\frac{D}{D_i} = 1 - 2.17 \cdot \frac{W_e}{W_c} \left(\frac{a}{R} \right)^2$$

$$\cong 1 - 1.085 \cdot \frac{\rho_e}{\rho_c t} a^2 \left(\frac{1}{R} \right) \quad (2)$$

where, ρ is density, and the subscripts e and c respectively denote the explosive and the confinement. The size effect of the detonation velocity has been discussed by many researchers using the relationship between the detonation velocity and the inverse radius. Although the two above models have been reconsidered and modified, since the discussion using the inverse radius

is still meaningful, the investigation of our experimental data will start using curved-front theory.

3. Results and discussion

Figure 5 shows the size effect of an AN using the relationship between the detonation velocity and the inverse radius. Equation (2) becomes invalid in the case of an extremely thin wall. Even so, when considering steel tubes with various thicknesses, it is very useful to remove the effect caused by differences in thickness. One of the authors previously investigated the size effect of AN using large-diameter steel tubes^{22),25)}, the results of which are also plotted in Figure 5. The solid line in Figure 5 was drawn using Equation (2) with the average reaction zone length obtained from the data. The results for the large-diameter case are well reproduced by Equation (2). This shows that the assumption of a spherical wave at the detonation front approximately holds in the large-diameter case, and that the detonation velocity at an infinite charge diameter can be predicted from experimental data by fitting. In contrast, in the case of a small diameter, the fitting line shown by the dashed line in Figure 5 does not attain the value at an infinite charge diameter. This means that curved-front theory cannot be applied to estimate the detonation velocity at an infinite radius by fitting in the case of a small diameter. Since the ideal detonation velocity for each set of data was calculated by the Cheetah code, it is possible to summarize the data using Equation (2). To attain the detonation velocity at an infinite charge diameter, a dramatic change in the reaction zone length is required according to curved-front theory, meaning that comparison of the theory and the experimental data requires three different fitting lines (the dotted lines in Figure 5). The parameter a in Equations (1) and (2) was considered to be the reaction zone length in the 1940s. Later, the existence of an effective C-J state in the highly non-ideal explosive was suggested and supported. Since

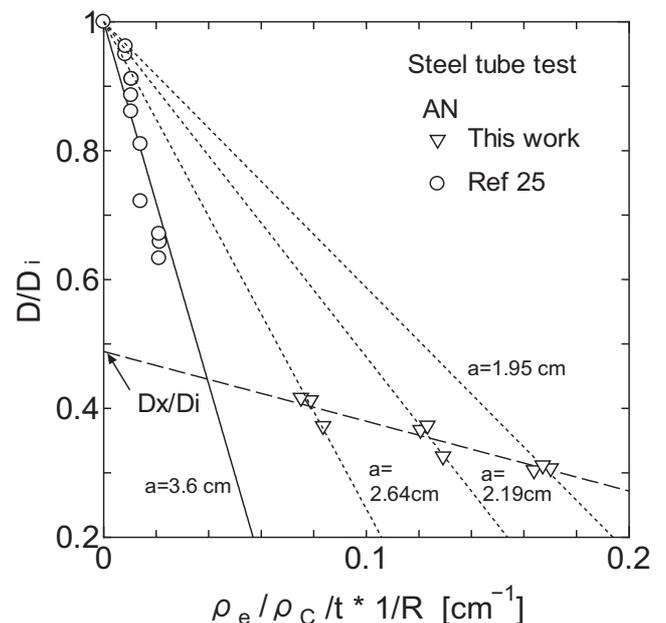


Figure 5 Size effect curve for ammonium nitrate with comparison of the curved-front theory.

the effective C-J state has similar characteristics to the C-J state, if the assumption of a spherical detonation front is still valid, Equation (2) may be adopted to predict the length of the effective reaction zone. Souers *et al.* proposed a model of the size effect and used the length of the effective reaction zone instead of the reaction zone length^{8),12)}. The effective reaction zone is defined by the distance from the detonation front to the sonic plane in the reaction zone. The parameter a decreased with decreasing charge diameter as shown in Figure 5. It can be considered that, together with the decreasing in the diameter, the effect of the wall of the steel pipe becomes sufficiently for the detonation front to approaches the plane wave. This situation can generate a detonation wave with the effective driving zone in the reaction zone. There is a specific diameter at which all of the detonation energy is supplied to sustain the detonation propagation. This suggests that above this diameter, the slope of the size effect curve changes.

Although the theoretical background has to be investigated in future, in the region mainly governed by the confinement effect, the characteristic line for the size effect can be expressed as

$$\frac{D}{D_i} = D_x - 1.083 \cdot \frac{\rho_e}{\rho_c t} \left(\frac{1}{R} \right) \quad (3)$$

This empirical equation has a non dimensional parameter D_x at $1/R = 0$ as shown in Figure 5. Figures 6 and 7 show the size effect curves for AN and AN/AC mixtures. Except for the tube with 20.4mm inner diameter, all the experimental results satisfy Equation(3). In the case of adding from 0.1 to 5 wt% AC, the reaction energy, which contributes to the detonation propagation increased with increasing AC. This tendency has already been shown in Figure 2. Figures 6 and 7 show that the same tendency exists for all charge diameters. When the mixing ratio of AC is λ , the detonation velocity of AN/AC mixture for $0.1 < \lambda < 5.0$ can be defined by Equation(3) using the fitting equation

$$D_x(\lambda) = 0.7187\lambda^{0.1477} \quad (4)$$

For a mixing ratio of 1.0 wt% AC, as shown in Figure 7, the discrepancy between the prediction line and the experimental data is confirmed for small diameters. It is considered that for small diameters, since the generated pressure is relatively low, the effect of the stiffness of the steel pipe on the reaction rate cannot be ignored. This problem will be considered in a future study on non-ideal detonation.

4. Conclusion

A series of steel tube tests on AN/AC mixtures have been conducted. The non-ideal behaviour of pure AN in a steel tube was examined by considering the relationship between the detonation velocity and charge radius using curved-front theory. Although in the case of a large radius, the detonation velocity at an infinite charge diameter could be estimated directly from the experimental data, it

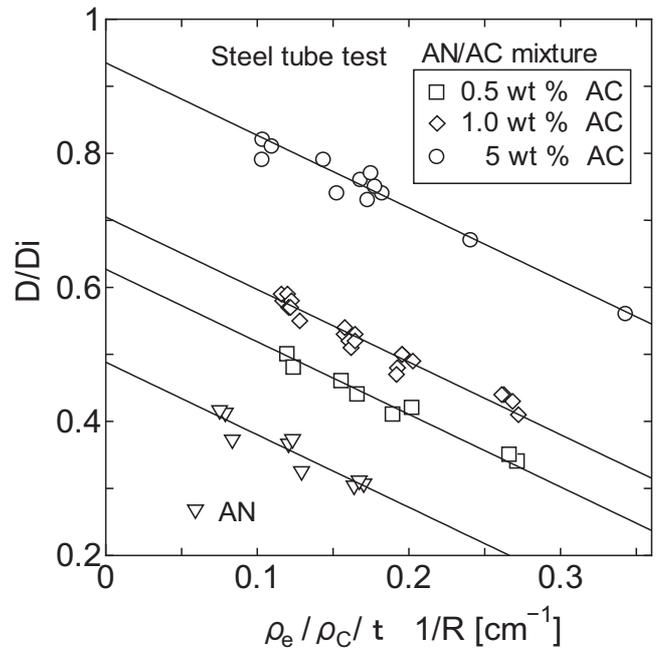


Figure 6 Size effect curve for ammonium nitrate and mixture of the ammonium nitrate and activated carbon (0.5 wt%, 1.0 wt%, and 5 wt% activated carbon).

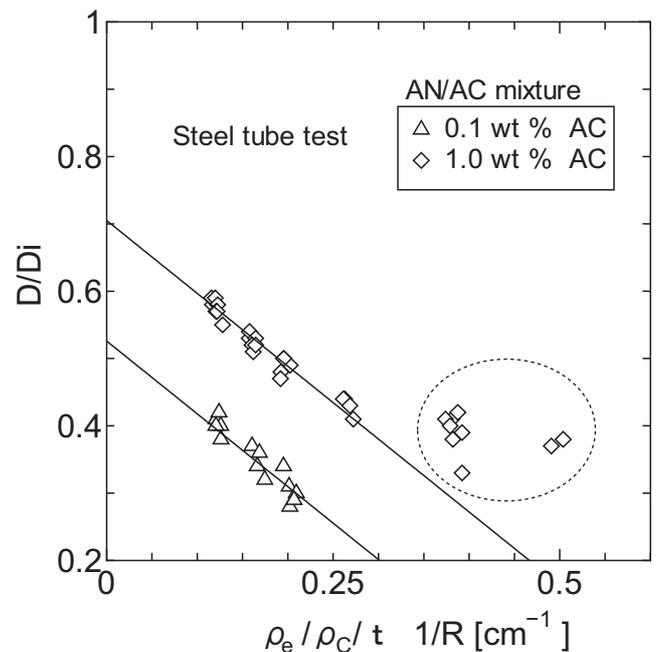


Figure 7 Size effect curve for ammonium nitrate and mixture of the ammonium nitrate and activated carbon (0.1 wt%, and 1.0 wt% activated carbon).

could not be predicted using the experimental data obtained from tubes with a relatively small radius in this study. The size effect curve for pure AN has a point of inflection, and most of it can be approximately expressed by two straight lines. Finally, the size effect curves for AN/AC mixtures were constructed and formulated for almost the entire experimental region.

References

- 1) H. Jones, Proc. Roy. Soc. (London), A189, 415–426 (1947)
- 2) H. Eyring, R. E. Powell, G.H. Duffey, and R.B. Parlin, Chem.

- Rev., 45, 69–181 (1949)
- 3) W. W. Wood, and J. G. Kirkwood, *J. Chem Phys.*, 21, 1920–1924 (1954).
 - 4) L. D. Sawin, R. H. Stresau, S. J. Porter, and J. Savitt, Third symposium on (international) detonation, 309–325, Princeton, New Jersey (1960).
 - 5) H. Eyring, F. E. Walker, S. Ma, and N. Coon, *Poc. Natl. Acad. Sci. USA*, 77, 2358–2361 (1980).
 - 6) C. L. Mader, “Numerical Modeling of Detonations”, (1979), University of California Press
 - 7) J. B. Dzil, and D. S. Stewart, *Phys. Fluids A*, 1., 7, 1261–1267 (1989).
 - 8) P. C. Souers, ‘Propellants Explos. Pyrotech.’, 22, 221–225 (1997)
 - 9) P. C. Souers, S. Anderson, J. Mercer, E. McGuire, and P. Vitello, ‘Propellants Explos. Pyrotech.’, 25, 54–58 (2000).
 - 10) R. A. Catanach, , and L. G. Hill, Shock Compression of Condensed Matter –2001, 906–909, Atlanta, Georgia (2001).
 - 11) A. Maranda, A. Paplinski, D. Galzowski, *Energetic Materials*, 21, 1–3 (2003).
 - 12) P. C. Souers, P. Vitello, S. Esen, J. Kruttschnitt, and H. A. Bilgin, ‘Propellants Explos. Pyrotech.’, 29, 19–26 (2004).
 - 13) S. Esen, *Rock Mech. Rock Engn.*, 37, 317–330 (2004).
 - 14) H. R. James, B. D. Lambourn, C. A. Handley, N. J. Whitworth, H. N. Angseeing, P. J. Haskins, M. D. Cook, A. D. Wood, R. I. Briggs, and P. R. Ottley, Thirteenth international detonation symposium, 110–120, Norfolk, Virginia (2006).
 - 15) B. Zygumt, and D. Buczkowski, ‘Propellants Explos. Pyrotech.’, 32, 411–414 (2007).
 - 16) B. L. Wescott, “Generalized pseudo-reaction zone model for non-ideal explosives”, Shock Compression of Condensed Matter –2007, 433–436, Waikoloa, Hawaii (2007).
 - 17) A. E. Medvedev, V. M. Fomin, and A. Yu. Reshetnyak, *Shock waves* 18, 107–115 (2008).
 - 18) S. I. Jackson, C. B. Kiyanda, and M. Short, “Precursor detonation wave development in ANFO due to Aluminum confinement”, Fourteenth international detonation symposium, 740–749, Coeur d’Alene, Idaho (2010).
 - 19) T. R. Salyer, M. Short, C. B. Kiyanda, J. S. Morris, and T. Zimmerly, “Effect of prill structure on detonation performance of ANFO”, Fourteenth international detonation symposium, 758–768, Coeur d’Alene, Idaho (2010).
 - 20) M. Short, J. J. Quirk, C. B. Kiyanda, S. I. Jackson, M. E. Briggs, and M. A. Shinas, “Simulation of of detonation of ammonium nitrate fuel oil mixture confined by aluminum : Edge angles for DSD”, Fourteenth international detonation symposium, 769–778, Coeur d’Alene, Idaho (2010).
 - 21) A. Miyake, H. Kobayashi, H. Ehigoya, S. Kubota, Y. Wada, Y. Ogata, H. Arai, and T. Ogawa, ‘Journal of Loss Prevention in the Process Industries’, 20, 548–588 (2007).
 - 22) A. C. van der Steen, H. H. Kodde, and A. Miyake, ‘Propellants Expos. Pyrotech.’, 15, 58–61 (1990).
 - 23) A. Miyake, K. Takahara, T. Ogawa, Y. Ogata, Y. Wada, and H. Arai, ‘Journal of Loss Prevention in the Process Industries’, 14, 533–538 (2001).
 - 24) A. Miyake, K. Takahara, T. Ogawa, Y. Ogata, and H. Arai, *Kayaku Gakkaishi (Sci. Tech. Energetic Materials)*, 63, 279–282 (2002).
 - 25) A. Miyake, A. C. van der Steen, and H. H. Kodde, “Detonation velocity and pressure of the non-ideal explosive ammonium nitrate”, Proc.9th Int’l Symposium on Detonation, 560–565, Portland (1989).
 - 26) A. Miyake, N. Kinoshita, S. Kubota, T. Saburi and Y. Ogata, “Non ideal detonation of ammonium nitrate and activated carbon mixtures in steel pipes”, Fourteenth international detonation symposium, 1246–1251, Coeur d’Alene, Idaho (2010).
 - 27) N. Kinoshita, S. Kubota, T. Saburi, Y. Ogata, and A. Miyake, *Sci. Tech. Energetic Materials*, 72, 21–25 (2011)
 - 28) L. E. Fried, W. M. Howard, and P. C. Souers, “Cheetah 2.0 user’s manual”. USA: Lawrence Livermore National Laboratory, (1998).

硝安系高エネルギー物質の小径鋼管内における 非理想爆轟挙動

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寸法効果曲線は爆轟速度と薬径の逆数の関係で表せる。この論文で、我々は硝安系爆薬の鋼管内爆轟試験によって得られた寸法効果曲線について議論した。硝安ならびに硝安／活性炭混合物の結果が調査された。活性炭の混合比は0.1～20 wt%，鋼管の内径は5.7～65.9 mmの範囲で変化させ、その爆轟速度への影響を検討した。純粋な硝安の寸法効果曲線は変曲点を持ち2つの直線で表現できることが判明した。さらに、硝安／活性炭混合物の寸法効果曲線を定式化した。

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