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Nonideal detonation regimes in low density explosives

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Measurements using Velocity Interferometer System for Any Reflector (VISAR) were performed for three high explosives at densities slightly above the natural loose-packed densities. The velocity histories at the explosive/window interface demonstrate that the grain size of the explosives plays an important role. Fine-grained materials produced rather smooth records with reduced von Neumann spike amplitudes. For commercial coarse-grained specimens, the chemical spike (if detectable) was more pronounced. This difference can be explained as a manifestation of partial burn up. In fine-grained explosives, which are more sensitive, the reaction can proceed partly within the compression front, which leads to a lower initial shock amplitude. The reaction zone was shorter in fine-grained materials because of higher density of hot spots. The noise level was generally higher for the coarse-grained explosives, which is a natural stochastic effect of the highly non-uniform flow of the heterogeneous medium. These results correlate with our previous data of electrical conductivity diagnostics. Instead of the classical Zel'dovich–von Neumann–Döring profiles, violent oscillations around the Chapman–Jouguet level were observed in about half of the shots using coarse-grained materials. We suggest that these unusual records may point to a different detonation wave propagation mechanism. © 2016 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4942359>]

I. INTRODUCTION

In the 1940s, Zel'dovich,¹ von Neumann,² and Döring³ (ZND) developed a hydrodynamical theory of detonation waves. Since then, the ZND model has become the predominant paradigm in detonation studies. In its primary form, the detonation wave is considered to be a complex that consists of (1) a leading shock that ignites the chemical reaction; (2) a reaction zone, in which the flow is subsonic, thus making it possible to support the leading shock; (3) the sonic Chapman–Jouguet (CJ) plane where the reaction is completed; and (4) a supersonic non-stationary Taylor expansion wave.⁴ This theory, which was certainly an outstanding intellectual achievement, was accepted by the scientific community well before any experimental confirmation. Evidence of the von Neumann spike, first reported in 1955 by Duff and Houston,⁵ as well as many subsequent results, still boosted the status of the ZND model.

It was later found that the detonation process is much more complicated in many cases. For instance, a gaseous detonation might be expected to provide an ideal example of the standard ZND case. However, instead of a plane shock followed by a chemical reaction zone, a complex flow, known as spin or cellular detonation, is formed, in which a substantial area of the leading shock is too weak to start the reaction and the main energy release that supports the process occurs in the stronger transverse waves. Nevertheless, the primary propagation agent is a shock wave, so in a certain broader sense, the ZND ideology is justified.

Surprisingly, even an analog of the CJ plane can be discussed, which ensures the stable wave velocity.⁶ Thus, the ZND model has proven to be quite robust. At present, detonation is thought to be ZND process, and the initial and simplest form of the theory is typically assumed.

It is commonly held that for solid explosives, the ZND theory works well enough. Although the leading shock usually cannot induce a homogeneous reaction, the explosive is ignited in multiple “hot spots.” The hot spots originate from structural defects, e.g., pores or cracks, that are always present in real solid explosive charges. The averaged effect of those reaction sites produces the classic wave profile with a von Neumann spike. Current reaction flow models, e.g., the ignition and growth model of Tarver *et al.*,⁷ are based on the ZND framework.

Historically, exactly solid explosives provoked certain doubts in the applicability of the ZND model. The natural alternative was some sort of convective process in which hot reaction gases penetrate through the pores to the fresh explosive particles, causing their ignition. The pressure gradient supports the gas flow as well as the fragmentation of the initial particles, which enables high reaction rates. This concept was advocated by Apin,⁸ for example, and was referred to as “explosive burning.” Apin even suggested that this mechanism is valid for high density explosives and attributed the convective heat transfer to micro-cumulative jets that formed in residual pores or in microscopic low-density defects. The convective contribution to the detonation propagation was also discussed by Guirguis *et al.*⁹ who used the term “convective detonation.”

Although Apin's extension of the idea to high-density explosives is probably an extremity, explosive burning in

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low density solid explosives a priori appears to be a reasonable mechanism of detonation propagation. Actually, the ZND concept does require a certain mental effort to be applied to loose packed materials in which the explosive grains occupy about the same fraction of the total volume as the air-filled pores. For this kind of structure, it is difficult to imagine a leading shock (although not impossible, one has to assume an immediate homogenization of the material, for example). Alternatively, if one accepts that the average pressure front is smeared over the initial explosive grain size and that the characteristic time required to crush the grains is at least approximately the same as the pressure rise time, then random jets that force their way from the high-pressure region would be a natural phenomenon. Thus, the actual working mechanism should be determined experimentally. Due to the current respectable status of the ZND model, any deviation from it would be of interest.

These considerations have prompted several researchers to study low density explosive systems. Solov'ev *et al.*¹⁰ prepared a special material based on hexogen (RDX) grains distributed in a plastic sponge, with a low average explosive density of 0.2–0.8 g/cm³. It was found that below 0.6–0.8 g/cm³, the reaction propagated due to the flow of the detonation products. Above ≈ 1.1 g/cm³, the leading shock was presumably the main propagation agent, and at 0.8–1.1 g/cm³, both mechanisms were competing. Pinaev,¹¹ who used still lower concentrations of explosive dust distributed in a rigid porous structure, also reported non-ZND convective processes.

For dilute systems, in which the explosive grains divided by gaps cannot transmit pressure waves, it is easy to agree with the explosive burning mechanism. However, at high densities, the ZND model is well justified. The situation for the intermediate case, i.e., loose packed explosives with densities of approximately 1 g/cm³, is both controversial and interesting. Unfortunately, there are few works that cover this density range. This can be readily explained, as explosives at high densities are of greater practical interest. Moreover, for low density explosives, due to their porous structures, one can expect ragged experimental records which are not a good stimulus for immediate study. For example, in the paper of Tarver *et al.*,⁷ the velocity history of the interface between the pressed pentaerythritol tetranitrate (PETN) and the LiF window was recorded using 100 nm thick gold layer. This inertia-free mirror provided a nanosecond time resolution. At the same time, it could reflect the laser light over approximately 1 μ s because of perfect homogeneity of the explosive, which was pressed to a high density of 1.76 g/cm³ (1% porosity). Typical low density charges with porosities of 30%–40% would immediately destroy such a thin film. Thus, for a long time, the nearly singular reference source that treated a wide range of densities was the book by Dremine *et al.*,¹² who found no deviations from the ZND model at densities of 1 g/cm³. However, these data were obtained using an early electromagnetic technique, and their time resolution (100 ns) was rather poor. Therefore, certain revisions to their conclusions would be appropriate. Later, a wide range of explosive densities was covered by Utkin *et al.*^{13,14} who used the Velocity Interferometer System for Any Reflector (VISAR) method with rather thick

foils to reduce the effects of turbulence caused by the porous structures of explosive charges. Their idea was that the foils mainly transmit the wave, perhaps introducing some attenuation but not qualitatively distorting its profile. Unfortunately, although thick foils provide smooth records, they may conceal fine details because of the averaging of the flow velocity. Additionally, short peaks can be blurred during the wave transmission, and the reaction zone of the explosive might be affected by the shock reflected from the thick Al layer. Thus, the fundamental difficulty caused by the heterogeneous structure of the porous charges was not completely overcome because foils less than 50 μ m thick were found to be unsuitable for this technique.

Nevertheless, Utkin *et al.* were able to demonstrate reproducible von Neumann peaks that were 30 to 40 ns in duration, which is several times longer than the transmission time in 50 μ m foil. Thus, their results deserve attention. An important fact is that qualitatively different profiles were obtained for the same explosive depending on the density and the grain size. In many cases, Utkin *et al.* found either reduced chemical spikes (presumably due to partial reaction within the shock) or even inverted wave structures with increasing, rather than decreasing, pressure. This latter case was explained as a manifestation of either dissipative processes in the detonation front or of the underdriven detonation. Although the first explanation seems unlikely, the underdriven regime was actually demonstrated¹⁴ in trinitroethyl trinitrobutyrate, or TNETB (an explosive similar to RDX). In the same range of porosity (3.3% to 12.5%) in which the inverted profiles were obtained, the detonation velocity was distinctly higher than that interpolated between “normal” density intervals. Let us remind that the underdriven regime, being faster than the normal Chapman–Jouguet wave, cannot propagate owing to the leading shock.⁴

In an important work by Gustavsen *et al.*,¹⁵ porous octogen (HMX) at a density of 1.24 g/cm³ (35% porosity) was studied using electromagnetic gauges. In fine-grained material (10 μ m grain size), a wave profile of reasonable quality was obtained, and the maximal pressure was midway between an unreacted shock and a Chapman–Jouguet state. In coarse-grained HMX (120 μ m), the peak “just barely reached the estimated CJ point,” and considerable noise was observed, with a signal to noise ratio of about 1. The “coarse” profiles were not reproducible. These observations point to a certainly non-ZND flow type. The role of cumulative jets formed in cavities within the charge, which are capable of penetrating into the unreacted explosive, was stressed by Fedorov.¹⁶

In our recent paper,¹⁷ the electrical conductivity profiles behind the detonation fronts in low density HMX, RDX, and PETN were measured. We found that the conducting zones in the coarse-grained explosives were approximately two times wider than in fine-grained ones. This difference is attributed to higher reaction rates in the fine-grained explosives, which clearly had denser populations of hot spots. The coarse grained explosives also produced higher noise levels, although less pronounced than that found by Gustavsen *et al.*¹⁵ Bearing in mind these results, we found it promising to study the structure of the detonation waves in the same low density explosives using conventional VISAR flow

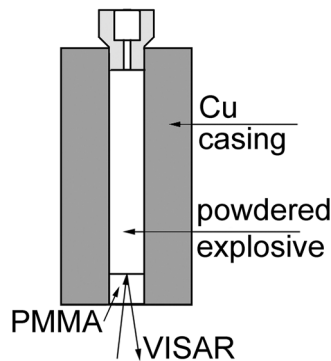


FIG. 1. Schematic of the experimental cell. The detonation wave propagates downwards.

diagnostics. Previously, we applied this method to highly heterogeneous emulsion explosives,¹⁸ in which the voids, namely, the interiors of glass microballoons, may occupy an even larger part of the total volume than the pores in loose packed explosives.

II. EXPERIMENTAL SETUP

A sketch of the experimental cell is shown in Figure 1. Except for the measuring technique, all of the conditions resembled those used in the tests¹⁷ to make the subsequent comparison easier. An explosive charge (8 mm in diameter) was placed in a copper casing that had 30 mm outer diameter. The copper shell served in the previous work¹⁷ as an outer electrode of a coaxial system. The detonation was initiated through an axial channel (2.4 mm in diameter) drilled in the plastic stopper. The charge length was greater than 50 mm, i.e., it was long enough to form a stationary process. The end face of the charge contacted with the PMMA window through a 100 μm thick epoxy layer and 7 μm of Al foil. Because the shock response of epoxy is close to that of PMMA, the protecting epoxy layer was supposed to transmit the pressure wave to the foil without serious distortion. Thicknesses of 100 μm usually provided a microsecond lifetime for the foil used in the present study, as well as in our experiments with emulsion explosives.¹⁸

The velocity history of the foil was recorded using a VISAR laser Doppler interferometer (VMBV-04 type from Valyn VIP, Inc.) with a constant of 1050 m/s per interference fringe. In most of the shots, the detonation velocity was measured using a special contact gauge.

The explosives tested were HMX, RDX, and PETN. The densities were slightly higher than those of natural fillings to remove macroscopic blobs and voids. Two kinds of each material were used: a common coarse grained dispersion and a

TABLE I. Characteristics of explosives.

| Explosive | HMX | RDX | PETN |
|-----------------------------------|-----|-----|------|
| d (coarse), μm | 430 | 160 | 260 |
| d (fine), μm | 21 | 11 | 80 |
| ρ_0 , g/cm^3 | 1.3 | 1.2 | 1.1 |
| τ (coarse), ns | 56 | 69 | 92 |
| τ (fine), ns | 31 | 34 | 59 |

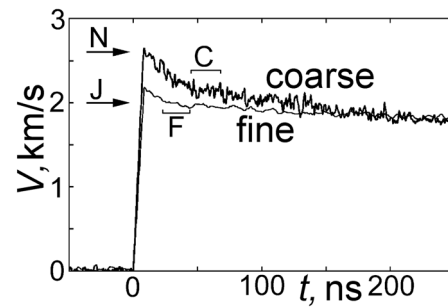


FIG. 2. Interface velocity V vs time t for RDX.

fine-grained one. The materials were the same as those used in the electrical conductivity study.¹⁷ The median grain sizes d , initial densities ρ_0 , and average durations of the conductivity peaks τ are listed in Table I.

III. RESULTS

In Figure 2, two records obtained with coarse- and fine-grained RDX are compared. The fine-grained material produces much smoother profiles, which is natural because the spot (say 1 mm in diameter) of the laser light on the foil covers several thousands of the 11 μm grains. Thus, the inevitably turbulent character of the flow is sufficiently averaged. Moreover, the velocity distribution of various sections of the foil is smoothed by the epoxy layer, which is rather thick compared to the explosive grain size. On the contrary, the same spot covers several tens of the larger 160 μm grains. Hence, the statistical averaging and smoothing are less pronounced. A similar dependence of the noise on the grain size was reported by Gustavsen *et al.*¹⁵ The results were generally reproducible for both the coarse-grained and fine-grained RDX.

To estimate the theoretical interface velocity that should be induced by the explosive, we used the PMMA shock Hugoniot fit¹⁹ (the relation between the shock velocity U_S and the particle velocity U_P is $U_S = 2.59 + 1.52U_P$ km/s) and the explosive equation of state calculations of Tanaka *et al.*²⁰ The latter reference is convenient because it contains data for a broad range of initial densities that can be easily interpolated to our conditions. The arrow J in Figure 2 points to the interface velocity generated by the CJ state of RDX at the initial density of 1.2 g/cm^3 . The arrow N points to the expected velocity level that might be produced by the unreacted (von Neumann) peak state. Because this state is not known with certainty, we used the simplest estimate by assuming that the pressure and the particle velocity in the shock point are 40% higher than that in the CJ state. For RDX and PETN, the von Neumann states defined in this way fall practically onto the PMMA shock Hugoniot, so there was no need to recalculate the corresponding interface velocities. For HMX, the correction was less than 150 m/s. Note that the peak value of the “fine” profile is between the CJ and von Neumann states, as found by Gustavsen *et al.*¹⁵ and, in several cases, by Utkin *et al.*^{13,14} However, the peak in the coarse-grained material is close to the von Neumann level.

The horizontal segment F in Figure 2 marks the range in the durations of the electrical conductivity peaks measured

for the fine-grained RDX.¹⁷ There is a rather good agreement between this conduction time and the reaction time, which can be roughly estimated as the moment at which the velocity reaches the Chapman–Jouguet level (i.e., the intersection of the extension of the J arrow and the velocity profile). The horizontal segment C marks the durations of the conductivity peak in the coarse-grained RDX. Here, the reaction zone seems to be somewhat longer, although there is also a qualitative agreement. Note that the durations of the conductivity peaks¹⁷ are Eulerian values, whereas in the present work, the measured times are close to Lagrangian values which are greater in proportion to the degree of compression in the detonation wave for approximately 30%–40%. That correction would improve the agreement between the results of the two diagnostics.

The reduced value of the peak interface velocity may be understood as a manifestation of the partial burnout of the explosive within the shock. Utkin *et al.*¹⁴ observed that the shock in a highly porous system is not an instantaneous jump but a layer of complex random flow of finite average thickness (say, one grain diameter), in which the material is converted from a two-phase gas-particle mixture to a more or less homogeneous medium. In highly porous explosives, several processes may produce ignition in the hot spots, namely, Mach stems that are generated in pores or jets that impact the solid grains, in addition to collapsing pores that are more relevant to dense explosives. Thus, a chemical reaction may partly proceed within the said layer. The lower level of velocity in the fine-grained explosive presumably means that the degree of conversion reached within the front is high enough to be visible. Because the density of the hot spots is higher in the fine-grained material, the reaction should be faster behind the shock layer as well, which can be seen in Figure 2.

In Figure 3(a), the interface velocity histories for the two kinds of HMX are presented. As in the RDX case, the “fine” records are smoother and reproducible, and their peaks are between the CJ and von Neumann values. One difference is that the “coarse” peak happened to be slightly higher than the estimate of the von Neumann state (probably because that estimate is too crude). The lack of reproducibility is more important. In Figure 3(b), two shots with the coarse-grained HMX are compared.

The second shot demonstrates considerable noise, and the average signal is close to the CJ level. This latter case is similar to the corresponding result of Gustavsen *et al.*¹⁵ Because the grain size of the coarse HMX is comparable to the diameter of the light spot, the signal is generated by the area that is in contact with several grains at most, and with little, if any, smoothing in the epoxy layer, which explains the noise. The velocity history is interrupted at 200 ns because of the foil’s destruction and/or the loss of contrast (see below). The characteristic period of the oscillations roughly corresponds to the ratio of the grain diameter d to the detonation velocity D (approximately 70 ns). In this connection, the good, classical shape of the first “coarse” record seems to be a lucky accident. If a pocket of smaller grains was assembled around the illuminated area or if the large pores nearest to the light spot were filled with the smaller

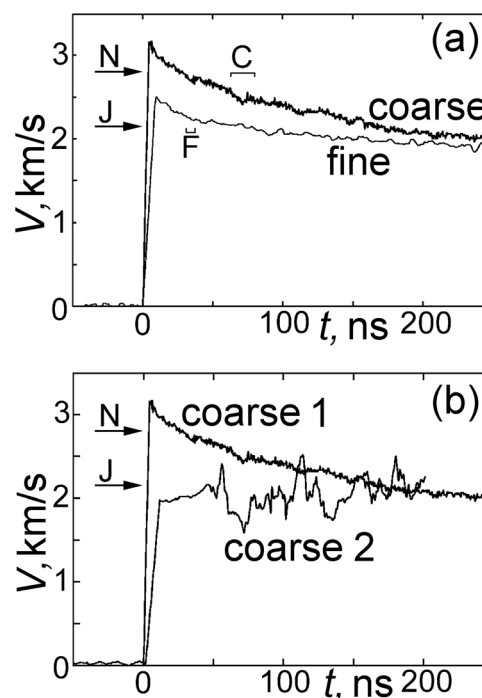


FIG. 3. Interface velocity for HMX. (a) comparison of the profiles for the coarse-grained and fine-grained materials. (b) Comparison of the profiles for two coarse-grained shots. The notation is the same as that in Figure 2.

grains, one should expect a smoother profile. Micrographs of the explosive specimens¹⁷ show that a noticeable fraction of the HMX particles are smaller than average RDX grains. Unfortunately, we could not control the charge structure at the interface.

Similar to the RDX case, there is rather good agreement between the reaction time and the conductivity peak time for the fine-grained HMX. For the coarse-grained HMX, we either have a poorer correlation (within approximately a binary order of magnitude in Figure 3(a)) or the reaction time cannot be estimated at all (as for the second profile in Figure 3(b)).

The results for PETN are shown in Figure 4. Again the peak amplitude in the fine-grained material is intermediate between the expected CJ and von Neumann velocities, and the conductivity time is close to the estimated reaction time.

As for the coarse-grained PETN, violent oscillations are observed around the Chapman–Jouguet level, or perhaps around a slightly higher one. Thus, the reaction time cannot

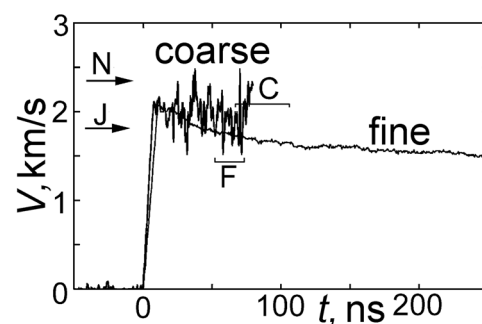


FIG. 4. Interface velocity for PETN. The notation is the same as that in Fig. 2.

be found. The record is interrupted after 80 ns, presumably because the foil was destroyed. This interruption and the form of the profile point to a highly turbulent flow character. Note that the noise frequency is much higher than in HMX case. The characteristic period is about 10 ns although the d/D ratio (50 ns) was almost the same as for HMX. The second shot with coarse-grained PETN gave a similar result, i.e., intense fast oscillations around the CJ level and the rapid destruction of the foil.

It should be noted that the common procedure of extracting the interface velocity from the VISAR data assumes that the foil surface moves uniformly within the area irradiated by the laser. If the foil is driven by random pulses applied at multiple places, the signal is a superposition of reflections from various spots moving with different velocities. Obviously, this will produce more complex and noisier signals. In this connection, Asay and Barker²¹ noted that the most serious problem is the loss of fringe contrast, which may prevent recovering the velocity. For the narrow velocity distribution, they demonstrated that the average velocity and even typical variations can be extracted from VISAR data. Our experiments with coarse-grained explosives present an opposite case of a broad velocity distribution. Although an average velocity can be found with some confidence, the computed oscillations may differ from the real ones. Nevertheless, we suppose that the oscillations in the velocity profiles maintain a certain resemblance to the natural velocity field, at least for the most general aspects such as the characteristic amplitude and the period of the pulsations. In particular, the different frequencies found for HMX and PETN suggest that diverse processes occur in these explosives.

IV. DISCUSSION

In the fine-grained low density explosives, the wave structure is in general agreement with the ZND model corrected for the partial conversion within the wave front. The latter should be thought of as a finite zone that is approximately one grain size thick. The duration of the front is several nanoseconds for grain sizes of 10–20 μm and could not be resolved in our experiments. The partial reaction in the front as well as the faster reaction behind it agrees with our observation¹⁷ that the reaction zone is shorter in fine-grained explosives. A good agreement is observed between the reaction times estimated from the VISAR profiles and those found from the electrical conductivity measurements.

However, in the coarse-grained materials, the experimental records may be either of ZND type (as in RDX and sometimes HMX) or of a different sort (as in PETN and sometimes HMX). The latter flow type is extremely chaotic. Because the characteristic time d/D for coarse-grained explosives may be of the same order of magnitude as the total chemical reaction time, the reaction zone is an essentially two-phase (gas and solid) flow. Hence, gaseous jets should arise ahead of the average wave front. However, although these jets are certainly present merely because of the structure of the material, they may not necessarily determine the wave propagation. If the explosive is insensitive to

the hot gas flow filtering through the pores, if the grains are easily crushed into tiny fragments that block the filtration or if the stress propagates from one particle to its neighbor effectively through their contact areas, then the reaction may be ignited either within the grains or in the Mach stems in the pores due to the overall compression, and the main propagation factor is the pressure wave. This presumably takes place in RDX. Alternatively, if the material is especially sensitive to the hot gas jets, the jets can become the main propagation agents. One may speculate that PETN is an example of such a material. The high-frequency oscillations of the velocity profile may be caused by the fast random ignition of the explosive ahead of the average wave front. HMX seems to be an intermediate case in which the jets and flow turbulence may be visible but are usually not intense enough to ignite the explosive. If the possibility of a convective mechanism, also called an explosive burning, is admitted, transitional variants may also exist, e.g., the detonation might switch from the shock-driven to convective mode and vice versa in the same charge.

In our experience,¹⁸ in emulsion explosives, the velocity profiles take the classic ZND shape, despite the high volume fraction of voids. The reason why the convective processes have a lesser effect in that case is the extremely low sensitivity of the reactive component of the emulsion explosives. Indeed, an ammonium nitrate and fuel oil mixture usually cannot detonate without the addition of microballoons. The latter play the role of artificial hot spots, in which the reaction is ignited as a result of their collapse. Of course, cumulative jets may form during the compression of the voids, but they cannot propagate the reaction ahead of the main wave front. On the contrary, PETN (in which a non-ZND process is probable) is one of the most sensitive secondary explosives according to most testing procedures.

We realize that the results and arguments presented above do not prove that the convective mechanism has actually been found. However, there seems to be certain evidence that it is a possibility. It would be interesting to find independent confirmations, although, at present, it is difficult to propose any direct and decisive method.

At present, low density explosives are unpopular among researchers who use modern high resolution methods. One reason for this neglect is the noisy form of the signals that are expected from such materials. Profiles resembling those in Figures 3(b) and 4 might be obtained from time to time, but these results most probably would be thrown into the recycle bin. The paper of Gustavsen *et al.*¹⁵ is a rare example of consistency. We would like to stress that the low density range is an interesting field especially because non-ZND regimes may be expected.

ACKNOWLEDGMENTS

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