

# Hybrid Detonation Waves

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## Introduction

While fine aluminum particles suspended in air are not sensitive to detonation due to a large transverse wave spacing, their combustion in gaseous detonation products may support so called hybrid detonation and hybrid deflagration-to-detonation transition (DDT). Afanasieva et al. (1983) postulated the existence of “double-shock” combustion in multiphase media using a one-dimensional flow theory. Veyssiere (1984) reported for the first time the observation of a detonation wave comprised of a double-shock in a lean reactive gas mixture with aluminum particles at concentrations ranging 15-80 g/m<sup>3</sup>. The test was conducted in a 6 m long apparatus consisting of a 4.5 m long by 6.9 cm diameter circular tube connected to a 5.3 cm square tube. Using a two-phase ZND model, Khasainov and Veyssiere (1988) stated that a “steady” double-shock detonation structure can exist, in which the two fronts are stabilized by the generalized CJ condition for the particle-gas mixture. Recent experiments have provided more conclusive evidence on the steady double-shock detonation and explored other hybrid detonation modes (Zhang et al. 2003, 2004). DDT in hybrid mixtures with large concentrations of aluminum particles near a tube end wall was also reported to result in a peak pressure of more than twice that produced in the same gas system alone (Zhang et al. 1998). This was attributed to the reverberating shock initiation dynamics in a dense combustible particle layer. In the present paper, the one-dimensional theory has been applied to obtain the solutions of various hybrid detonation modes, in particular, the weak detonation solutions supported by the particle reaction. Based on the theoretical prediction, experimental work has also been continued to investigate the hybrid weak detonation modes in a lab-scale two-phase detonation tube for a range of initiation energy, size and concentration of micrometric grades of aluminum particles, oxidant composition of the host gas detonation products, and initial pressure.

## One-Dimensional Theory

Apart from the chemical non-equilibrium process, detonation of a solid particle-fluid mixture comprises other non-equilibrium processes of mass, momentum and energy transfer between the two phases due to finite sizes of solid particles. In general, the non-equilibrium mass, momentum and energy transfer depend on the physical properties of the particles and therefore do not have the same relaxation length scales as that of the chemical non-equilibrium process. Hence, a steady detonation solution cannot be achieved a priori without integration along the reaction path to determine the mechanical and thermal partial-equilibrium between the two phases. A one-dimensional two-phase ZND model has been introduced with a generalized C-J condition as a rear boundary condition, where the net heat release rate resulting from the chemical reaction and inter-phase mass, momentum and heat transfer approaches zero at the

gaseous frozen sonic locus (Lee & Sichel 1986; Khasainov & Veyssiere 1988, 1995; Fan & Sichel 1988).

From the two-phase, steady, one-dimensional conservation equations written in the coordinate frame with respect to the leading shock front propagating at velocity  $D$ , one can obtain a system of ordinary differential equations in which

$$\frac{du_l}{dx} = \Psi / \eta \quad (1)$$

with  $u_l$  to being the gas-phase flow velocity. Here

$$\eta = 1 - u_l^2 / c_{fl}^2 \quad (2)$$

is a sonic parameter of the flow with respect to the gaseous frozen sound speed  $c_{fl}$ . The quantity  $\Psi$  represents the “thermicity”, a measure of the rate of transformation from net energy release of all non-equilibrium processes to molecular and bulk translational energy. A “generalized multiphase C-J condition” serves as the rear boundary condition at the gaseous frozen sonic point imbedded in the reaction zone by finding the common zeros of the thermicity  $\Psi$  and the sonic parameter  $\eta$ :

$$\Psi = 0 \text{ at } \eta = 0. \quad (3)$$

The detailed expression of the thermicity depends on the non-equilibrium processes and equations of state. To elucidate the physical meaning of the total heat release rate, an analytical expression is given below for a simple system comprising a perfect gas with single exothermic (heat  $q_1 > 0$ ), irreversible gaseous reaction (rate  $w_1 > 0$ ), and a negligible volume fraction of solid particles ( $\phi_p = 0$ ) with exothermic particle combustion (heat  $q_p > 0$ , *inter-phase mass transfer*  $J_p > 0$ ) and constant particle number in the system:

$$\begin{aligned} \Psi = & \frac{\gamma - 1}{\rho_l c_{fl}^2} \left\{ q_1 w_1 + \sum_{p=2}^N (q_p + c_p T_p) J_p \right. \\ & - \sum_{p=2}^N \left[ (u_p - \frac{\gamma u_l}{\gamma - 1}) f_p + Q_p + \frac{(u_l - u_p)}{2} (u_p - \frac{(\gamma + 1) u_l}{\gamma - 1}) J_p \right] \\ & \left. + (\frac{\gamma u_l}{\gamma - 1} - D) f_w + Q_w \right\}. \end{aligned} \quad (4)$$

The first term on the right hand side of (4) describes the energy release rate of the gas phase reaction. The second term (with source term  $J_p$ ) represents the rate of energy release in the gas due to particle evaporation and reaction. The third term (in square brackets with inter-phase mass, momentum and energy transfer source terms  $J_p$ ,  $f_p$  and  $Q_p$ ) corresponds to the rate of gas energy change caused by the non-equilibrium flow velocity and temperature between the two

phases. The last two terms (with the “w” subscript) represent the rate of gas energy change due to momentum and heat transfer,  $f_w$  and  $Q_w$ , to lateral boundaries.

The ordinary differential equation system deduced from the conservation equations, the equations of state for the particles and gas phase, together with the generalized C-J condition (3), form the closure of the mathematical description of the two-phase ZND model, given the source terms for the exchange between the two phases and to the lateral boundaries. Thus, under the initial conditions of the post-shock state, a steady solution can be obtained for the velocity and reaction zone structure of the detonation wave in a solid particle-fluid system. If the activity energy of the reactive system is low, the steady solution can also be obtained by the long-time asymptotic solution of the one-dimensional unsteady conservation equations, which further proves the validity of the steady solution based on the generalized C-J condition. It is noticeable that the lateral boundary source terms,  $f_w$  and  $Q_w$ , only change solution values but are not necessary in satisfying the multiphase generalized C-J condition and the steady solution. For clarity, examples in this paper mainly address the multiphase effects and, therefore, do not include lateral boundary source terms.

The generalized C-J locus determined by (3) is a mathematical saddle point, after which the subsonic flow relative to the shock front can become supersonic as it reaches the weak detonation branch of the final equilibrium Hugoniot curve. The solution of the weak detonation depends on the physical and chemical properties of a reactive system and the boundary conditions behind the gas reaction zone. Two important conditions must be met for a steady weak detonation solution as follows.

1. The necessary conditions are: i) within the reaction zone, there is at least one gaseous frozen sonic point imbedded at which the generalized C-J condition is satisfied; and ii) the final equilibrium Hugoniot is not the upper bound of all partial-equilibrium Hugoniot curves.
2. The uniqueness of a steady weak detonation solution depends on the flow or boundary conditions behind the generalized C-J point.

A simple illustrative example of the weak detonation solution is the detonation wave in a perfect gas with an irreversible exothermic reaction followed by a secondary irreversible endothermic reaction, where heat releases  $|q_1| > |q_2|$  (Fickett & Davis 1979). It has a saddle point featured with the generalized C-J condition (3) imbedded in the reaction zone due to the endothermic reaction rate competing with the exothermic reaction rate. Thus, a steady solution can be obtained by integration of the ZND model from the post-shock state downstream to satisfy the generalized C-J condition at which the Rayleigh line is tangent to a partial-equilibrium Hugoniot curve. The partial equilibrium Hugoniot corresponds to the highest attainable heat release of the system,  $Q_{max} = q_1 + q_2(1 - e^{q_1/q_2})$ , that is larger than the heat release  $Q_f = q_1 + q_2$  in the final equilibrium Hugoniot. Therefore, the solution satisfies the necessary conditions for a weak detonation. It is then possible to continue the integration from the saddle point downstream, as the flow smoothly transits from subsonic to supersonic until it meets the final equilibrium Hugoniot curve. The detonation velocity  $D_m$  corresponding to the Rayleigh line tangent to the  $Q_{max}$ -Hugoniot is greater than the final equilibrium detonation velocity  $D_{CJ}$ . Depending on the

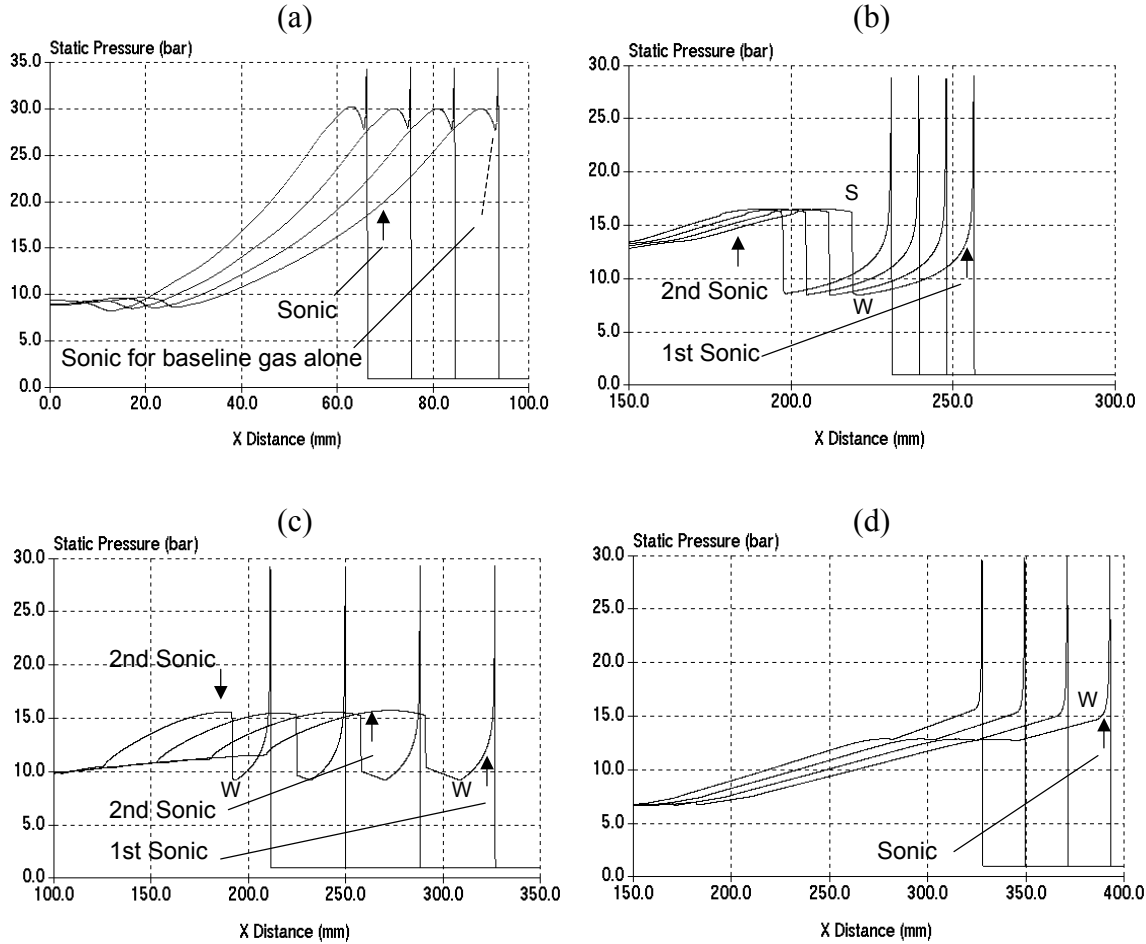
rear flow or boundary conditions, the solution can either return to a strong detonation point along the  $D_m$ -Rayleigh line or down to a weak detonation point where the flow is supersonic. The solution is incomplete without taking into account the second condition stated above. A variety of weak detonation solutions can be obtained when the detonation wave is followed by a piston of specified constant velocity by adjusting the piston velocity.

For a solid particle-gas mixture, a partial equilibrium state also includes that of mass, momentum and energy transfer processes between the two phases. The necessary conditions for a steady weak solution can be satisfied in some particle-gas flow cases by examining the thermicity (4) and partial-equilibrium Hugoniot curves. In fact, detonation in an inert particle-reactive gas system is analogous to that of the aforementioned two irreversible reaction gas system, where the role of the second endothermic reaction is fulfilled by the gas-phase momentum and heat loss to relatively large particles (Zhang et al. 1994). The inert-particle-reactive gas detonation has a saddle point imbedded in the reaction zone as depicted by the generalized C-J condition (3). Secondly, the final equilibrium Hugoniot lies below some partial-equilibrium Hugoniot curves because the final equilibrium detonation velocity is smaller than that of the partial-equilibrium Hugoniot curve for relatively large particles. After the gaseous sonic locus, the solid particle velocity and temperature will further equilibrate with that of the gas phase towards the final equilibrium Hugoniot as the flow becomes supersonic with respect to the leading shock front. The solution for weak detonation is incomplete without taking into account rear flow or rear boundary conditions.

Detonation in a reactive particle-reactive gas system is analogous to the detonation wave for the two irreversible reactions with the second reaction endothermic, followed by a piston of specified constant velocity. By choosing physical properties and flow variables for the solid particles, the momentum and heat loss from the gas to the particles can be regulated within the gas reaction zone to satisfy the necessary conditions for a weak detonation, while various weak detonation solutions can be realized by controlling the late particle reaction to meet rear flow conditions behind the saddle point. The heat release rate of particles can be adjusted through the particle material, size and concentration, or gaseous detonation parameters and products compositions. Assuming that the particle reaction follows the gas reaction, a set of steady solutions can be obtained through the long-time asymptotic solution of the unsteady conservation equations, as shown in Fig. 1. The characteristic parameter to specify a possible solution is the energy release rate of particles represented by the second term in the thermicity (4), denoted as  $q_p'J_p$ . The energy release rate of particles is assumed to have a time delay with respect to that of the gas phase reaction.

a) If  $q_p'J_p$  rises early and significantly, particle reaction can produce a compression wave in the gas reaction zone to increase detonation velocity and pressure (Fig. 1a). The entire subsonic reaction zone is substantially extended due to particle combustion and a steady solution is reached when the generalized C-J condition (3) is satisfied downstream at the gaseous frozen sonic locus. There exists a minimum in the pressure profile within the reaction zone when the net heat release reaches a local maximum at thermicity  $\Psi = 0$  before the sonic locus. This solution is referred to as “single-front detonation” by Veyssiere and Khasainov (1995), but was termed a “strong hybrid solution” (Zhang et al. 2003) in the sense that particle combustion within the reaction zone overdrives the gas detonation. This terminology came from the analogy with

detonation in the exothermic-endothermic two-reaction gas followed by a piston moving faster than the flow velocity of the strong detonation point. However, unlike the usual overdriven detonation where the entire flow is subsonic with respect to the leading shock front, a strong hybrid detonation will not be disturbed by the supersonic rear flow behind the sonic locus.



**Figure 1. Pressure-distance profiles for steady hybrid detonation solutions in a mixture of lean acetylene-air and aluminum particles: a) strong detonation,  $d_p = 2 \mu\text{m}$ ,  $\sigma_p = 300 \text{ g/m}^3$ ; b) type-I weak detonation,  $d_p = 10 \mu\text{m}$ ,  $\sigma_p = 500 \text{ g/m}^3$ ; c) type-II weak detonation,  $d_p = 20 \mu\text{m}$ ,  $\sigma_p = 500 \text{ g/m}^3$ ; and d) type-III weak detonation,  $d_p = 40 \mu\text{m}$ ,  $\sigma_p = 500 \text{ g/m}^3$ .**

b) When  $q_p'J_p$  is delayed to enable particle reaction behind the gas reaction zone, particles behave as though inert within the gas reaction zone and the necessary conditions for a steady weak detonation can be satisfied, where a generalized C-J point appears for the first time. In the supersonic gas flow behind the first sonic point, heat release from the particles would cause a continuous decrease in gas flow velocity to subsonic levels, and an increase in gas pressure. This, however, will not match the downstream unsteady supersonic flow required by the rear boundary condition, and instead result in thermal choking. Consequently, a second shock wave is necessary in order to adjust the gas flow behind the gas reaction zone from supersonic to

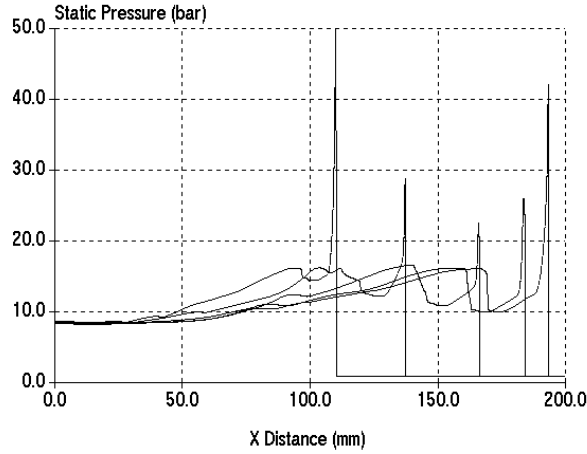
subsonic. The flow, with the heat release from the particles, is then able to expand towards the second sonic locus, where the generalized C-J condition is satisfied a second time to match the downstream unsteady supersonic flow. Thus, a double-shock solution can be achieved that consists of the steady gas reaction zone followed by a secondary shock. The  $q_p'J_p$ -induced second shock wave corresponds to a post-shock subsonic state  $S$  and a pre-shock supersonic point  $W$  behind the steady gas reaction zone. If the Rayleigh line  $SW$  coincides with that for the leading front, the second shock moves with the same velocity as the leading front (Fig. 1b). This solution is referred to as the type-I double-shock weak solution, analogous to the double-shock solution in the two-reaction gas followed by a piston velocity equal to the flow velocity of the strong detonation point.

c) When  $q_p'J_p$  decreases, the velocity of the  $q_p'J_p$ -induced second shock wave is reduced. The shock therefore recedes from the supersonic end state  $W$  of the steady gas reaction zone to produce an ever-widening region of supersonic flow between state  $W$  and itself (Fig. 1c). As  $q_p'J_p$  further decreases, the strength of the secondary shock decreases and recedes more rapidly. This solution is called the type-II double-shock weak solution, in analogy to the solution in the two-reaction gas followed by a piston with velocity between that of the strong detonation point and the weak detonation point. Unlike the weak detonation in the two-reaction gas followed by a piston, the ever-widening region of the supersonic flow is not uniform. The initial particle combustion increases the pressure and decreases the flow velocity upstream of the secondary shock. Furthermore, the particle reaction zone length between the second shock and the second sonic locus increases continuously as the shock recedes. Rigorously speaking, a steady solution does not exist after the end point  $W$  of the steady gas reaction zone.

d) As  $q_p'J_p$  is further delayed and reduced, the gas steady reaction zone end point  $W$  is connected to the supersonic rear flow imbedded with a weak compression wave caused by the particle combustion (Fig. 1d). While the detonation front propagates steadily and satisfies the generalized C-J condition (3) at the gaseous frozen sonic locus, the particle-reacting rear flow is unsteady and subject to the rear boundary condition. The solution is regarded as the type-III hybrid weak solution. The particles become chemically inert as  $q_p'J_p$  is reduced to approach a null value.

The one-dimensional multiphase ZND model has been found to be unstable (Zhang et al. 1994, 2003). For systems of any solid particles in reactive fluid, the momentum and heat transferred from the fluid to the particles within the fluid reaction zone destabilize the detonation. When the momentum and heat transferred to particles within the gas reaction zone exceed a limit, the steady hybrid weak detonation waves fail and unstable detonation with a transient secondary pressure wave can occur under conditions of appropriate particle heat release rate behind the gas reaction zone. Figure 2 illustrates a numerical simulation for a large concentration of 10  $\mu\text{m}$  aluminum particles suspended in a lean acetylene-air system. The unstable detonation wave propagates in an oscillatory mode at an average velocity 8% less than that of the baseline gas detonation, due to higher momentum and heat losses from the gas to the particles within the gas reaction zone. While the particles burn behind the gas reaction zone, the energy release from the particle combustion is coupled with the unsteady rear flow of the gas detonation. Consequently, a transient pressure wave is generated behind the detonation front. In an oscillatory cycle, the

pressure wave has an acceleration phase followed by a deceleration phase. The acceleration phase leads to the formation of a shock wave before the deceleration phase commences.



**Figure 2. Numerical simulation of unsteady weak hybrid detonation with a transient secondary pressure wave in a mixture of lean  $C_2H_2$ -air and  $1000 \text{ g/m}^3$ ,  $10 \text{ }\mu\text{m}$  aluminum particles.**

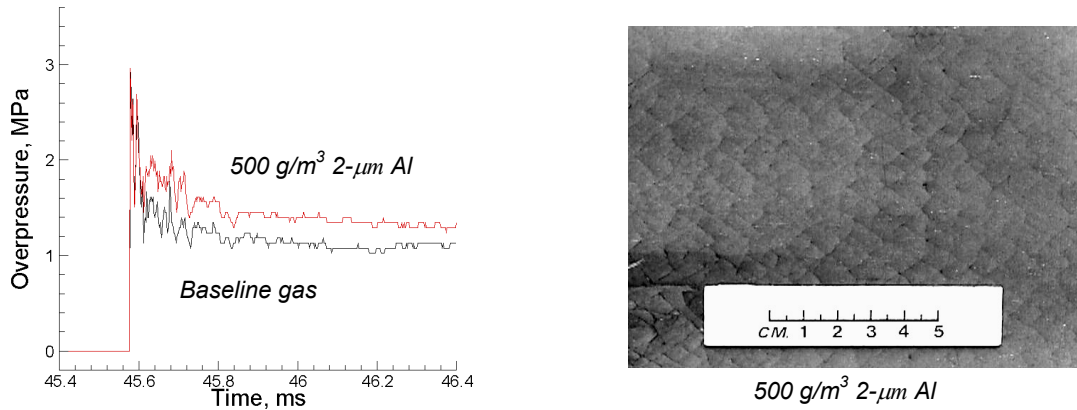
Although the structure of real detonation waves in solid particle-gas flow is transient and three-dimensional, the essence of the one-dimensional multiphase ZND model can never be over-addressed. Not only does it predict a mean structure averaged over the cross-section perpendicular to the propagation direction, but most importantly it provides a fundamental kernel of the unsteady, multi-dimensional continuum two-phase detonation theory.

## Experimental Results and Discussion

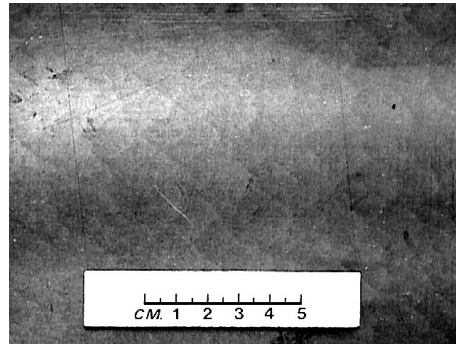
Experiments were conducted in a horizontal detonation tube of 80 mm internal diameter with a 10 m test section filled with a particles-gas suspension and a 3.1 m relief section. The initiation pulse was produced by a detonator (0.05-5 g) installed at the beginning of the test section. Micrometric grades of atomized aluminum supplied by Valimet Incorporated were tested and their sizes ranged from 2 to  $30 \text{ }\mu\text{m}$ . In order to examine the effect of various gaseous detonation products on aluminum combustion and the formation of two-shock hybrid detonation, acetylene-air, hydrogen-air, carbon monoxide-air and ethylene-air were chosen at various equivalence ratios and 1-2.5 atm initial pressures with detonation cell sizes ranging between 4 and 38 mm.

Three most important steady hybrid detonation modes are shown here in aluminum particle-gas mixtures with a particle concentration ranging from 250 to  $2000 \text{ g/m}^3$ . In all three modes, a pressure wave or shock wave was formed in the gas detonation flow due to particle combustion, thus enhancing the impulse loading. Figure 3 displays a steady strong hybrid detonation in  $2 \text{ }\mu\text{m}$  spherical aluminum particles suspended in  $C_2H_2$ -air. The strong hybrid detonation is characterized by the leading shock front followed by a compression wave in the gas detonation zone, caused by sufficiently large heat release rate of the small particles within the gas reaction

zone. This decreases the detonation cell size (Fig. 4) and increases the detonation velocity and pressure with respect to the baseline gas detonation alone, and therefore overdrives the gas detonation. The secondary compression wave and its inclusion in the gas detonation zone are more clearly resolved through a numerical simulation, as shown in Fig. 1a, where the compression wave penetrates two thirds of the detonation zone for the baseline case of gas alone. Recent experimental results of Veyssiere and Ingignoli (2003) also indicated that the mean detonation cell size can be reduced by 38 % and the detonation velocity is increased by 4 % when adding flaked aluminium particles to a lean hydrogen-air mixture.



**Figure 3. Strong hybrid detonation characterized by the first shock followed by a pressure wave in the gas reaction zone. Sample mixtures contain 2  $\mu\text{m}$  Al and  $\text{C}_2\text{H}_2$ -air ( $\phi = 0.8$ ) at 1 atm.**

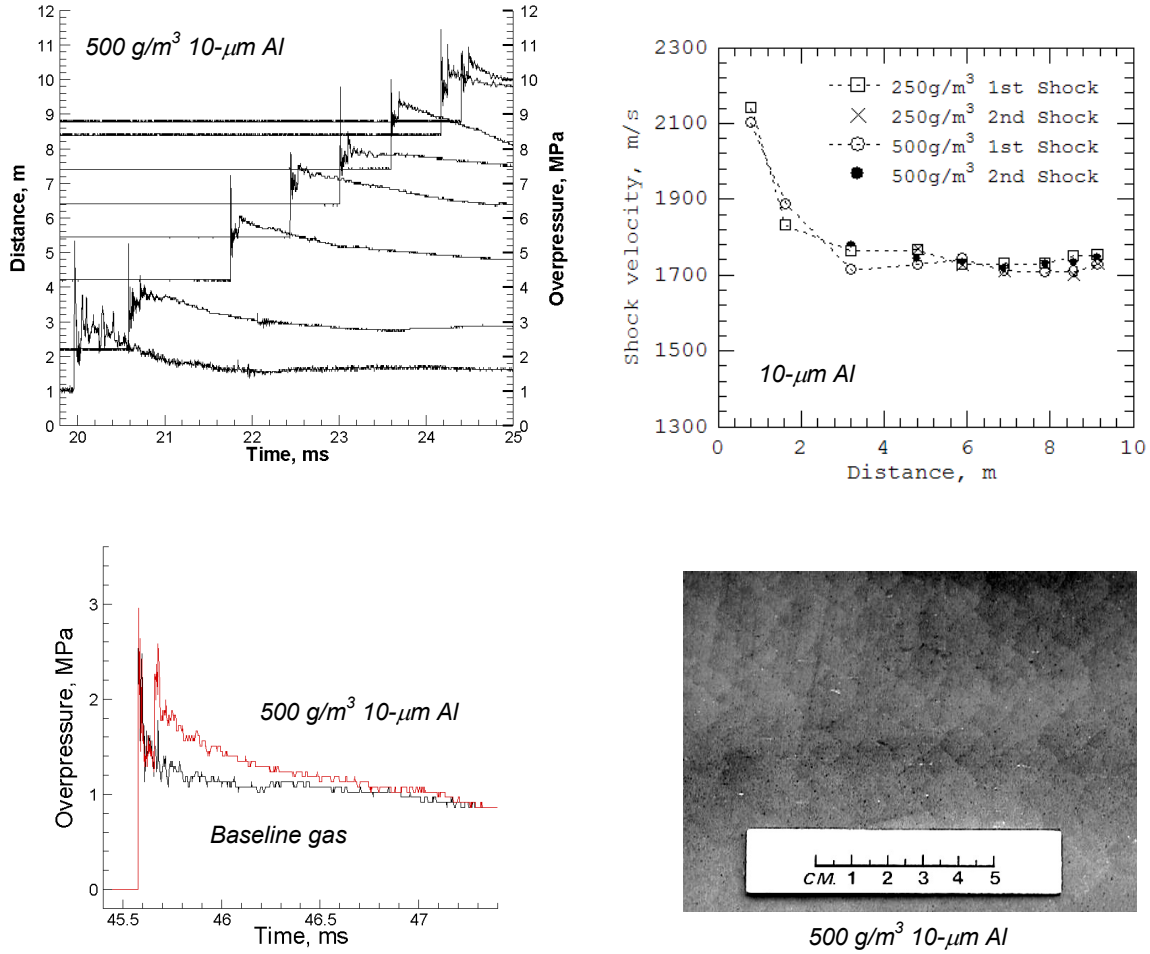


**Figure 4. Cellular detonation structure for detonation in the  $\text{C}_2\text{H}_2$ -air ( $\phi = 0.8$ ) baseline mixture at 1 atm.**

When the aluminum particle size was increased to 10  $\mu\text{m}$ , the particles behaved as inert within the gas reaction zone and particle heat release took place after the gas reaction zone. Therefore, a steady hybrid weak detonation resulted as shown in Fig. 5. This is the type-I double-shock weak solution characterized by a two-shock structure, where the second shock behind the

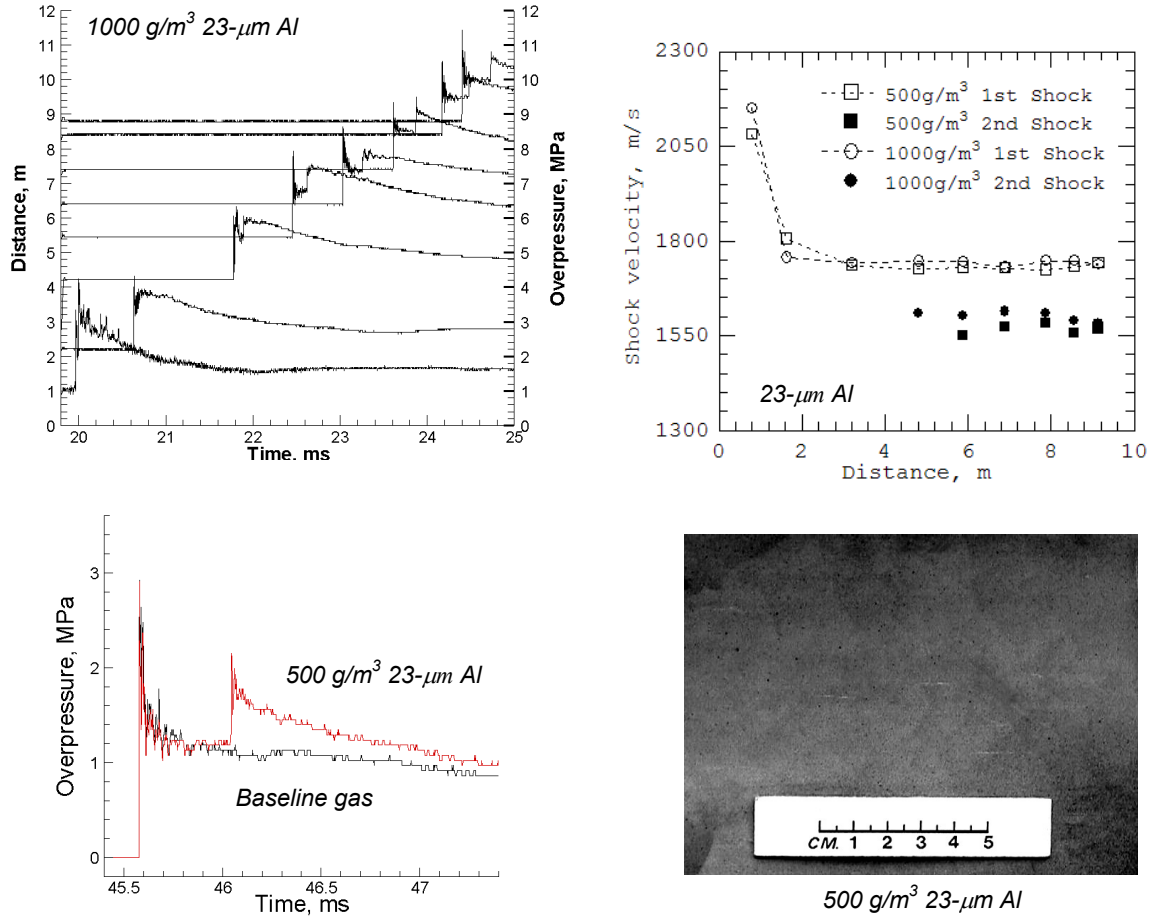


gaseous frozen sonic plane has the same velocity as the leading shock. The apparent detonation cell size printed on the smoke foil is mainly governed by the gas detonation due to insignificant momentum and heat transferred to the large particles within the gas detonation zone. Hence, this cell size does not provide information about the sensitivity of aluminum ignition and combustion.



**Figure 5.** Type-I weak hybrid detonation characterized by two shock fronts where the 2nd shock behind the gas reaction zone has the same velocity as the first. Sample mixtures contain 10  $\mu\text{m}$  Al and  $\text{C}_2\text{H}_2$ -air ( $\phi = 0.8$ ) at 1 atm.

On further increasing the aluminum particle size to 23  $\mu\text{m}$  to reduce the heat release rate of the particles, a type-II double-shock weak detonation was observed. This is characterized by two shock fronts, where the second front behind the gaseous frozen sonic plane has a velocity less than the leading front, as demonstrated in Fig. 6. Hence, the second shock recedes from the gas reaction zone to produce an ever-widening region of supersonic flow between the end of gas reaction zone and itself. Again, the detonation cell size recorded on the smoke foil apparently corresponds to that of the gas detonation due to the large particles used.



**Figure 6. Type-II weak hybrid detonation characterized by two shock fronts where the 2nd shock behind the gas reaction zone has a velocity less than the first. Sample mixtures contain  $23 \mu\text{m}$  Al and  $\text{C}_2\text{H}_2\text{-air}$  ( $\phi = 0.8$ ) at 1 atm.**

The influence of the gaseous detonation parameters and products composition on the steady hybrid detonation was investigated through systematic experiments in detonation of various equivalence ratios of acetylene-air, hydrogen-air, sensitized carbon monoxide-air and ethylene-air. It was found that the strong hybrid detonation and the two types of double-shock weak detonation waves can propagate steadily in detonation products with the presence of oxygen, water vapor or carbon dioxide, but propagation is unlikely in detonation products dominated by carbon monoxide. The experiments further justified the one-dimensional theory that the double-shock weak detonation waves strongly depend on the combination of the momentum and heat transferred to the particles within the gas reaction zone and the heat release rate of particles behind the gas reaction zone. By decreasing the heat release rate of particles through an increase in particle size or changing the products composition and parameters of gaseous detonation, the type-I double-shock weak detonation was shifted to the type-II, in which the second shock recedes with a weaker shock strength with a further decrease in the heat release rate of the

particles. An increase in initial pressure resulted in an increase in the heat release rate of the particles and therefore facilitated the steady hybrid detonation waves.

It is noticeable that the fuel-air gas detonation alone may not be sufficient to initiate combustion of relatively large particles quickly enough to form a secondary shock (Zhang et al. 2004). It was found that the secondary shock did not appear in the same particle-gas system with 23  $\mu\text{m}$  aluminum particles shown in Fig. 6 until using 5 g condensed charge initiation, while the  $\text{C}_2\text{H}_2$ -air detonation was already initiated directly and propagating steadily under a 0.2 g charge mass. These results clearly indicate that the fuel-air detonation itself is insufficient to initiate and maintain a hybrid double-shock weak detonation wave involving large aluminum particles and that additional initiation charges are required.

## Conclusion

Hybrid detonation waves occur in reactive particles suspended in a detonable gas. A variety of hybrid detonation modes can exist and the solution is a strong function of the gas reaction time scales and the additional time scales of the mass, momentum and heat transfer between the particles and the gas. According to the one-dimensional theory and systematic experimental results, a strong hybrid detonation wave and two types of double-shock weak detonation waves, among various modes, are most important in practice due to their enhancement to the gas detonation impulse loading. The strong hybrid detonation is characterized by the leading shock front followed by a compression wave resulting from particle combustion in the gas detonation zone, thus overdriving the gas detonation. The two types of double-shock weak detonation waves are featured by a two-shock structure where the second shock front caused by the particle combustion behind the gas reaction zone has a propagation velocity either the same as or less than the leading shock front, thus enhancing the impulse loading of the gas detonation. While the strong hybrid detonation reduces the cell size of the baseline gas detonation, the cell size in the hybrid weak detonation waves are mainly governed by the gas detonation and does not provide information about the sensitivity of aluminum ignition and combustion which often requires a strong initiation. The variety of hybrid detonation modes and their propagation mechanisms are still a subject of current research.

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