

On the behavior of detonation and deflagration waves in fluids with internal state variables

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In this paper we derive the differential equation which the amplitudes of detonation waves and deflagration waves propagating in chemically reacting mixtures must obey and examine the implications of this equation.

W niniejszej pracy wyprowadzono równanie różniczkowe, jakie muszą spełniać amplitudy fal detonacyjnych i fal wybuchowego spalania, rozprzestrzeniających się w chemicznie reagujących mieszaninach oraz przeanalizowano wnioski wypływające z tego równania.

В настоящей работе выведено дифференциальное уравнение, которому должны удовлетворять амплитуды детонационных волн и волн взрывного сгорания, распространяющиеся в химически реагирующих смесях, а также анализируются выводы вытекающие из этого уравнения.

1. Introduction

In this paper we consider the behavior of shock waves propagating in fluids with internal state variables. An example of such a fluid is a chemically reacting fluid mixture without diffusion for which the internal state variables represent the degrees of advancement of the various independent chemical reactions occurring in the mixture. The results given here differ from those in the literature in a fundamental way. Typically, in the analysis of shock waves in materials with internal state variables it is assumed that the state variables are continuous across the shock; but the derivatives of the state variables are allowed to have jump discontinuities. Such a model, when applied to reacting mixtures, allows the chemical reactions to take place only in the regions behind the shocks. Such shocks are said to be *frozen*.

In this paper we remove the frozen assumption and consider a class of stronger shocks in that we allow the internal state variables to have jump discontinuities. Physically, this means that abrupt changes in composition occur across the shock, and that the reaction zones are idealized as occurring at the shocks themselves. We also assume that immediately behind the shocks the fluid mixtures are in states of strong equilibrium. The precise definition of strong equilibrium is given in Sect. 3. For chemically reacting mixtures, the preceding assumptions correspond to the physical condition that the reactions induced by the shocks approach equilibrium infinitely fast.

Our objective in this paper is to examine the *growth and decay* properties of those shocks across which the internal state variables suffer jump discontinuities. The properties of the

instantaneous jumps across these shocks are well known. We believe that our results, which govern the evolution of these jumps in time, are new. From the standpoint of applications, our results are meaningful with regard to certain aspects of the behavior of shock waves in explosives. When the amplitudes of the shocks are small they will not induce any chemical reaction in the explosives and their behavior will be purely mechanical. However, when the amplitudes of the shocks are sufficiently large, chemical reactions are induced. It is well known that when this happens the amplitudes of the shocks will grow and become what are commonly called detonation waves. Our results describe the evolutionary behavior of detonation waves.

2. Preliminaries

In this paper, we consider the mathematical model of a non-conducting fluid with internal state variables. For this model the internal energy ε , the pressure π and the absolute temperature θ are determined by the specific volume v , the entropy density η and R internal state variables $\xi_1, \xi_2, \dots, \xi_R$. That is,

$$(2.1) \quad \varepsilon = \hat{\varepsilon}(v, \eta, \boldsymbol{\xi}), \quad \pi = \hat{\pi}(v, \eta, \boldsymbol{\xi}), \quad \theta = \hat{\theta}(v, \eta, \boldsymbol{\xi})$$

where $\boldsymbol{\xi}$ is the R -tuple $(\xi_1, \xi_2, \dots, \xi_R)$ and is called the internal state vector. The material derivative $\dot{\boldsymbol{\xi}}$ of $\boldsymbol{\xi}$ obeys the equation

$$(2.2) \quad \dot{\boldsymbol{\xi}} = \mathbf{h}(v, \eta, \boldsymbol{\xi}).$$

Also, it is well known that the Second Law of Thermodynamics requires ⁽¹⁾

$$(2.3) \quad \hat{\pi} = -\frac{\partial \hat{\varepsilon}}{\partial v}, \quad \hat{\theta} = \frac{\partial \hat{\varepsilon}}{\partial \eta},$$

$$(2.4) \quad \hat{\boldsymbol{\sigma}}(v, \eta, \boldsymbol{\xi}) \cdot \mathbf{h}(v, \eta, \boldsymbol{\xi}) \geq 0,$$

where

$$(2.5) \quad \boldsymbol{\sigma} = \hat{\boldsymbol{\sigma}}(v, \eta, \boldsymbol{\xi}) \equiv -\frac{\partial \hat{\varepsilon}(v, \eta, \boldsymbol{\xi})}{\partial \boldsymbol{\xi}}.$$

In the application of this model to a chemically reacting mixture without diffusion, $\boldsymbol{\xi}$ is the extent of reaction vector, $\dot{\boldsymbol{\xi}}$ is the reaction rate vector and $\boldsymbol{\sigma}$ is the chemical affinity vector.

In this paper, we consider the one-dimensional motions of the fluids characterized by (2.1) through (2.5). Such a motion is characterized by the function χ giving the position x at the time t of the material point X :

$$(2.6) \quad x = \chi(X, t).$$

Of course, we identify each material point with its position in a fixed reference configuration. The relation between the specific volume v and χ is simply

$$(2.7) \quad v = \frac{1}{\rho} = \frac{1}{\rho_0} \frac{\partial \chi}{\partial X},$$

where ρ is the present density and ρ_0 is the density in the reference configuration.

⁽¹⁾ Cf. COLEMAN and GURTIN [1], and BOWEN [2].

We assume that the motion contains a wave moving with velocity $U(t) = \frac{dY(t)}{dt} > 0$ where $Y(t)$ is the material point at which the wave is found at the time t . Let f denote ξ , v , ρ , \dot{x} or η . Then

(i) the motion χ is continuous everywhere;

(ii) f , \dot{f} and $\partial f/\partial X$ have jump discontinuities across the wave.

If ξ is continuous, then the wave is a *frozen shock wave*. The growth and decay properties of this type of shock wave were first investigated by CHEN and GURTIN [3]. The continuity of ξ follows from the assumption that there are no surface sources of mass at the wave. Here, we allow ξ to be discontinuous. This assumption means that as the wave passes through the fluid chemical reactions take place instantly⁽²⁾.

In view of (i) and (ii), it follows from (2.1) and (2.2) that ε , π , θ and $\dot{\xi}$ and their derivatives also have jump discontinuities across the wave. In addition, we have the following compatibility relations:

$$(2.8) \quad [\dot{x}] = -U \left[\frac{\partial \chi}{\partial X} \right] = -\rho_0 U [v],$$

and

$$(2.9) \quad \frac{d[f]}{dt} = [\dot{f}] + U \left[\frac{\partial f}{\partial X} \right],$$

where f denotes ξ , v , ρ , \dot{x} , η , ε , π or θ . Here, $[f] = f^- - f^+$ with $f^\mp = \lim_{X \rightarrow Y(t)^\mp} f(X, t)$.

Balance of linear momentum and balance of energy imply that

$$(2.10) \quad \rho_0 U [\dot{x}] = [\pi],$$

$$(2.11) \quad \rho_0 [\dot{x}] = - \left[\frac{\partial \pi}{\partial X} \right],$$

$$(2.12) \quad [\varepsilon] + \frac{1}{2} (\pi^- + \pi^+) [v] = 0,$$

$$(2.13) \quad [\dot{\varepsilon}] = - [\pi \dot{v}].$$

It follows from (2.2), (2.3) and (2.5) that formula (2.13) can be written in the more useful form

$$(2.14) \quad [\theta \dot{\eta}] = [\sigma \cdot \mathbf{h}].$$

Since we have taken $U(t) > 0$, the Second Law of Thermodynamics requires that

$$(2.15) \quad [\eta] \geq 0.$$

By (2.8), formula (2.10) yields the result

$$(2.16) \quad \rho_0^2 U^2 = - \frac{[\pi]}{[v]}$$

for the velocity of the wave. Here we should point out that, even though (2.16) has the usual form, the velocity may be quite different depending on how $[\xi]$ changes across the

⁽²⁾ Cf. R. COURANT and K. O. FRIEDRICHS [4].

wave. The jump $[v]$ is a measure of the strength of the wave. It can be shown that it must obey the kinematical formula⁽³⁾

$$(2.17) \quad 2U \frac{d[v]}{dt} + [v] \frac{dU}{dt} = U^2 \left[\frac{\partial v}{\partial X} \right] - \frac{1}{\rho_0} [\ddot{x}].$$

In the next section we utilize the preceding results to derive the differential equation which governs the growth and decay properties of $[v]$.

Finally, we assume, as is customary, that the following inequalities hold:

$$(2.18) \quad \frac{\partial \hat{\pi}(v, \eta, \xi)}{\partial v} < 0, \quad \frac{\partial \hat{\pi}(v, \eta, \xi)}{\partial \eta} > 0, \quad \frac{\partial^2 \hat{\pi}(v, \eta, \xi)}{\partial v^2} > 0.$$

3. The governing differential equation of the amplitude and its consequences

If we assume that $[\xi] \neq 0$ and formally follow the analysis given by CHEN and GURTIN [3], we may derive the governing equation of the amplitude of a wave propagating in a fluid characterized by the constitutive relations (2.1). However, now we are not able to deduce the implications of this equation because we have no knowledge as to how $[\xi]$ changes across the wave. Hence it seems that we must adopt some assumption which is physically reasonable and which yields the jump $[\xi]$ in terms of certain known properties of the wave.

We assume that the fluid ahead of the wave is initially in a state of *weak equilibrium* and that immediately behind the wave the fluid is in a state of *strong equilibrium*⁽⁴⁾. Physically, this means that the reactions induced by the wave approach equilibrium infinitely fast, and it is certainly consistent with our knowledge of the behavior of detonation and deflagration waves. This assumption is sometimes referred to as *shifting equilibrium*.

We assume that (2.5)₁ can be solved to yield

$$(3.1) \quad \xi = \hat{\xi}(v, \eta, \sigma)$$

for all (v, η) ; this in turn implies that the symmetric $R \times R$ matrix

$$(3.2) \quad \frac{\partial \hat{\sigma}(v, \eta, \xi)}{\partial \xi} = - \frac{\partial^2 \hat{e}(v, \eta, \xi)}{\partial \xi \partial \xi}$$

is non-singular. Given (3.1), we can write (2.2) in the form

$$(3.3) \quad \hat{\xi} = \mathbf{h}(v, \eta, \hat{\xi}(v, \eta, \sigma)) \equiv \hat{\mathbf{h}}(v, \eta, \sigma).$$

By (2.4) and the fact that (3.2) is non-singular, it is possible to show that it is necessary for

$$(3.4) \quad \hat{\mathbf{h}}(v, \eta, \mathbf{0}) = \mathbf{0}.$$

That is, when (3.1) holds and $\sigma = \mathbf{0}$, then the state must necessarily be a *strong equilibrium* state. When $\sigma = \mathbf{0}$, (3.1) may be rewritten in the form

$$(3.5) \quad \xi = \hat{\xi}(v, \eta, \mathbf{0}) \equiv \xi^e(v, \eta).$$

⁽³⁾ Cf. CHEN and GURTIN [3].

⁽⁴⁾ A weak equilibrium state is characterized by the triplet (v^+, η^+, ξ^+) where v^+ , η^+ , and ξ^+ are constants such that $\mathbf{h}(v^+, \eta^+, \xi^+) = \mathbf{0}$. In addition, if $\hat{\sigma}(v^+, \eta^+, \xi^+)$ is also zero, then the equilibrium state is said to be a strong one, cf. § IV of BOWEN [2], and Chap. 6 of TRUESDELL [5].

That is, the state vector ξ is determined by (v, η) .

By (3.5), we have

$$(3.6) \quad [\xi] = \xi^e(v^-, \eta^-) - \xi^+.$$

That is, $[\xi]$ is determined by $[v]$ and $[\eta]$.

Given (3.1), we see that in a state of strong equilibrium, we have

$$(3.7) \quad \varepsilon = \hat{\varepsilon}(v, \eta, \xi^e(v, \eta)) \equiv \varepsilon^e(v, \eta),$$

$$(3.8) \quad \pi = \hat{\pi}(v, \eta, \xi^e(v, \eta)) \equiv \pi^e(v, \eta),$$

$$(3.9) \quad \theta = \hat{\theta}(v, \eta, \xi^e(v, \eta)) \equiv \theta^e(v, \eta);$$

further, it follows from (3.7) and (2.5) that

$$(3.10) \quad \frac{\partial \varepsilon^e(v, \eta)}{\partial v} = \frac{\partial \hat{\varepsilon}(v, \eta, \xi^e(v, \eta))}{\partial v},$$

$$(3.11) \quad \frac{\partial \varepsilon^e(v, \eta)}{\partial \eta} = \frac{\partial \hat{\varepsilon}(v, \eta, \xi^e(v, \eta))}{\partial \eta}.$$

These results along with (2.3) yield

$$(3.12) \quad \pi^e = -\frac{\partial \varepsilon^e}{\partial v}, \quad \theta^e = \frac{\partial \varepsilon^e}{\partial \eta}.$$

Similarly, it follows from (2.18) that

$$(3.13) \quad \frac{\partial \pi^e(v, \eta)}{\partial v} < 0, \quad \frac{\partial \pi^e(v, \eta)}{\partial \eta} > 0, \quad \frac{\partial^2 \pi^e(v, \eta)}{\partial v^2} > 0.$$

Next, we utilize (2.11), (2.12), (2.14), (2.16), and (2.17) to derive the governing equation of the amplitude of the wave. An examination of these equations shows that the formal calculation is similar to that first given by CHEN and GURTIN [6] and CHEN [7] for shock waves in elastic non-conductors. Indeed, we have the following:

$$(3.14) \quad \frac{d[\eta]}{dt} = \frac{\left(\frac{\partial \pi^e}{\partial v}\right)^-}{\left(\frac{\partial \pi^e}{\partial \eta}\right)^-} \frac{\gamma^e(1-\mu^e)}{(2-\gamma^e)} \frac{d[v]}{dt},$$

$$(3.15) \quad \frac{dU}{dt} = \frac{U(1-\mu^e)}{\mu^e(2-\gamma^e)} \frac{d[v]}{dt},$$

$$(3.16) \quad \frac{d[v]}{dt} = -\frac{U(1-\mu^e)(2-\gamma^e)}{(3\mu^e+1)-\gamma^e(3\mu^e-1)} \left(\frac{\partial v}{\partial X}\right)^-,$$

where

$$(3.17) \quad \mu^e = -\frac{\rho_0^2 U^2}{\left(\frac{\partial \pi^e}{\partial v}\right)^-},$$

$$(3.18) \quad \gamma^e = -\frac{1}{(\theta^e)^-} [v] \left(\frac{\partial \pi^e}{\partial \eta}\right)^-.$$

In addition, it follows from (3.6) and (3.14) that

$$(3.19) \quad \frac{d[\xi]}{dt} = \left\{ \left(\frac{\partial \xi^e}{\partial v} \right)^- + \left(\frac{\partial \xi^e}{\partial \eta} \right)^- \frac{\left(\frac{\partial \pi^e}{\partial v} \right)^-}{\left(\frac{\partial \pi^e}{\partial \eta} \right)^-} \frac{\gamma^e(1-\mu^e)}{(2-\gamma^e)} \right\} \frac{d[v]}{dt}.$$

In the derivation of (3.14), (3.15), (3.16) and (3.19), we have assumed that $\gamma^e \neq 2$.

In order that we may interpret (3.16) we need to investigate more closely the jump conditions (2.12) and (2.16). Before we do this let us define what is meant by an exothermic wave. The wave is said to be exothermic if for the same specific volume and entropy the fluid immediately in front of the wave has a greater internal energy density or enthalpy density than the fluid immediately behind the wave⁽⁵⁾. For example,

$$(3.20) \quad \begin{aligned} \hat{\varepsilon}(v^+, \eta^+, \xi^+) &> \varepsilon^e(v^+, \eta^+), \\ \hat{\varepsilon}(v^-, \eta^-, \xi^+) &> \varepsilon^e(v^-, \eta^-). \end{aligned}$$

This definition together with the assumptions we have adopted allows us to quote directly the results given by COURANT and FRIEDRICHS [4], Chap. III, Part E. They showed that (2.12), (2.15) and (2.16) are satisfied by two types of waves: detonation waves and deflagration waves. For detonation waves the jump $[v]$ is negative; while for deflagration waves the jump $[v]$ is positive. In addition, for a detonation wave

$$(3.21) \quad \rho_0^2 U^2 > + \left(\frac{\partial \hat{\pi}}{\partial v} \right)^+;$$

while for a deflagration wave

$$(3.22) \quad \rho_0^2 U^2 < - \left(\frac{\partial \hat{\pi}}{\partial v} \right)^+.$$

Formula (3.21) states that the wave speed is supersonic with respect to the fluid in front of the wave. Formula (3.22) states that the wave speed is subsonic with respect to the fluid in front of the wave.

Now, there are two types of detonation waves ($[v] < 0$). The first type is called a *strong detonation wave* (SDe). It has the property

$$(3.23) \quad \rho_0^2 U^2 < - \left(\frac{\partial \pi^e}{\partial v} \right)^-.$$

That is, the wave speed is subsonic with respect to the fluid behind the wave. Another property of strong detonation wave is that the entropy jump $[\eta]$ increases with decreasing $[v]$, i.e.,

$$(3.24) \quad \frac{\partial [\eta]}{\partial [v]} < 0.$$

⁽⁵⁾ COURANT and FRIEDRICHS [4], p. 208.

The other type of detonation wave is called a *weak detonation wave* (WDe). In this case

$$(3.25) \quad \rho_0^2 U^2 > - \left(\frac{\partial \pi^e}{\partial v} \right)^-,$$

$$(3.26) \quad \frac{\partial [\eta]}{\partial [v]} > 0.$$

Of course, (3.25) states that the wave speed is supersonic with respect to the fluid behind the wave; and (3.26) states that the entropy jump $[\eta]$ decreases with decreasing $[v]$. The transition from a strong detonation wave to a weak detonation wave is smooth. The transition state has the properties

$$(3.27) \quad \rho_0^2 U^2 = - \left(\frac{\partial \pi^e}{\partial v} \right)^-,$$

$$(3.28) \quad \frac{\partial [\eta]}{\partial [v]} = 0.$$

The state for which (3.27) and (3.28) hold is called a Chapman-Jouguet detonation (C-JDe). For such a detonation, the wave speed is sonic with respect to the fluid behind the wave, and the entropy is a minimum relative to all the possible values it can attain for detonation waves.

There are also two types of deflagration waves ($[v] > 0$). A *weak deflagration wave* (WDf) has the properties

$$(3.29) \quad \rho_0^2 U^2 < - \left(\frac{\partial \pi^e}{\partial v} \right)^-$$

$$(3.30) \quad \frac{\partial [\eta]}{\partial [v]} > 0.$$

A *strong deflagration wave* (SDf) has the properties

$$(3.31) \quad \rho_0^2 U^2 > - \left(\frac{\partial \pi^e}{\partial v} \right)^-,$$

$$(3.32) \quad \frac{\partial [\eta]}{\partial [v]} < 0.$$

The transition from a weak deflagration wave to a strong deflagration wave is again smooth. The transition point is called a Chapman-Jouguet deflagration (C-JDf). At this state

$$(3.33) \quad \rho_0^2 U^2 = - \left(\frac{\partial \pi^e}{\partial v} \right)^-,$$

$$(3.34) \quad \frac{\partial [\eta]}{\partial [v]} = 0.$$

In summary, the wave speed is subsonic (sonic, supersonic) with respect to the fluid behind the wave when it is a weak (Chapman-Jouguet, strong) deflagration wave. In addition, the entropy jump $[\eta]$ increases with increasing $[v]$ for weak deflagration waves, reached a maximum at the Chapman-Jouguet state and then decreases with increasing $[v]$ for strong deflagration waves.

If we make use of the result

$$(3.35) \quad \frac{\partial[\eta]}{\partial[v]} = \frac{\left(\frac{\partial\pi^e}{\partial v}\right)^-}{\left(\frac{\partial\pi^e}{\partial\eta}\right)^-} \frac{\gamma^e(1-\mu^e)}{(2-\gamma^e)},$$

which follows from (3.14), then by (3.17), (3.18), (3.13)_{1,2} and the properties of detonation and deflagration waves, we can assign bounds to μ^e and γ^e . The results are as follows⁽⁶⁾:

	μ^e	γ^e
SDe	$0 < \mu^e < 1$	$0 < \gamma^e < 2$
C-JDe	$\mu^e = 1$	$0 < \gamma^e < 2$
WDe	$\mu^e > 1$	$0 < \gamma^e < 2$
WDf	$0 < \mu^e < 1$	$\gamma^e < 0$
C-JDf	$\mu^e = 1$	$\gamma^e < 0$
SDf	$\mu^e > 1$	$\gamma^e < 0$

We are now in a position to examine the implications of (3.16). Indeed, we have the following:

(i) Strong Detonation Waves

$$(3.36) \quad \frac{d|[v]|}{dt} < 0 \Leftrightarrow \left(\frac{\partial v}{\partial X}\right)^- < 0,$$

$$(3.37) \quad \frac{d|[v]|}{dt} > 0 \Leftrightarrow \left(\frac{\partial v}{\partial X}\right)^- > 0,$$

(ii) Chapman-Jouguet Detonation Waves

$$(3.38) \quad \frac{d[v]}{dt} = 0 \text{ for all } \left(\frac{\partial v}{\partial X}\right)^-$$

(iii) Weak Detonation Waves

(a) If $\frac{3\mu^e+1}{3\mu^e-1} > \gamma^e$, then

$$(3.39) \quad \frac{d|[v]|}{dt} < 0 \Leftrightarrow \left(\frac{\partial v}{\partial X}\right)^- < 0,$$

$$(3.40) \quad \frac{d|[v]|}{dt} > 0 \Leftrightarrow \left(\frac{\partial v}{\partial X}\right)^- > 0,$$

(b) If $\frac{3\mu^e+1}{3\mu^e-1} < \gamma^e$, then

$$(3.41) \quad \frac{d|[v]|}{dt} < 0 \Leftrightarrow \left(\frac{\partial v}{\partial X}\right)^- > 0,$$

⁽⁶⁾ The restriction $0 < \gamma^e < 2$ for C-JDe arises because of the following reasons. By (3.13)₂ and (3.18) we see that $\gamma^e > 0$. The case of $\gamma^e = 2$ is excluded in the derivation of (3.14), (3.15), (3.16) and (3.19). Hence by continuity with SDe and WDe, it follows that $0 < \gamma^e < 2$.

$$(3.42) \quad \frac{d[v]}{dt} > 0 \Leftrightarrow \left(\frac{\partial v}{\partial X} \right)^- < 0.$$

(iv) Weak Deflagration Waves

(a) If $\frac{3\mu^e + 1}{3\mu^e - 1} > \gamma^e$, then

$$(3.43) \quad \frac{d[v]}{dt} > 0 \Leftrightarrow \left(\frac{\partial v}{\partial X} \right)^- < 0,$$

$$(3.44) \quad \frac{d[v]}{dt} < 0 \Leftrightarrow \left(\frac{\partial v}{\partial X} \right)^- > 0,$$

(b) If $\frac{3\mu^e + 1}{3\mu^e - 1} < \gamma^e$, then

$$(3.45) \quad \frac{d[v]}{dt} > 0 \Leftrightarrow \left(\frac{\partial v}{\partial X} \right)^- > 0,$$

$$(3.46) \quad \frac{d[v]}{dt} < 0 \Leftrightarrow \left(\frac{\partial v}{\partial X} \right)^- < 0.$$

(v) Chapman-Jouguet Deflagration Waves

$$(3.47) \quad \frac{d[v]}{dt} = 0 \text{ for all } \left(\frac{\partial v}{\partial X} \right)^-.$$

(vi) Strong Deflagration Waves

$$(3.48) \quad \frac{d[v]}{dt} > 0 \Leftrightarrow \left(\frac{\partial v}{\partial X} \right)^- > 0,$$

$$(3.49) \quad \frac{d[v]}{dt} < 0 \Leftrightarrow \left(\frac{\partial v}{\partial X} \right)^- < 0.$$

Given (3.36) through (3.49), we can use (3.14) and (3.15) to examine the growth and decay properties of $[\eta]$ and U . If the derivatives $(\partial \xi^e / \partial v)^-$ and $(\partial \xi^e / \partial \eta)^-$ are known, then we can use (3.19) to examine the behavior of $d[\xi]/dt$.

In closing, we should point out that we have said nothing about the existence of detonation and deflagration waves. We have simply exhausted the various possibilities which might exist. The interested reader may consult COURANT and FRIEDRICHS [4], §§93-94, in which the question of existence is examined in detail.

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