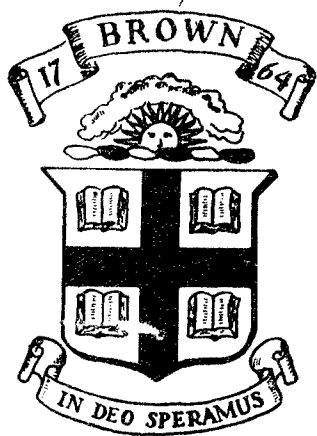
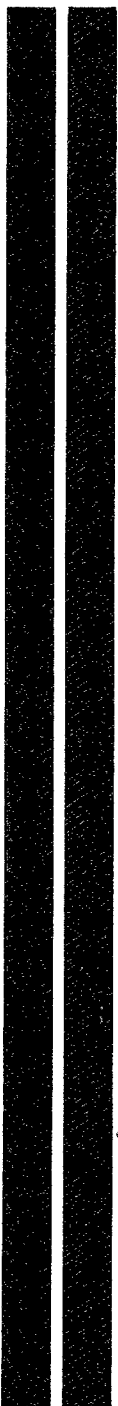


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Division of Engineering
BROWN UNIVERSITY
PROVIDENCE, R. I.



**A MACROSCOPIC THEORY OF
TWO-PHASE FLOW WITH MASS,
MOMENTUM AND ENERGY EXCHANGE
BY
BOA-TEH CHU AND J.Y. PARLANGE**

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U. S. Army Ordnance Corps
Picatinny Research Center, Picatinny Arsenal
Department of Army Project No. 5504-01-027
Ordnance R & D Project No. 755-5407N
Contract No. DA-19-020-ORD-4761

DA-4761/4

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Boa-Teh Chu and J. Y. Parlange

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under pp
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OCO, R and D Branch
(Project No. TS5-5407N)
Department of Army Project
No. 5S04-01-027

Technical Report No. 4

Division of Engineering
Brown University
Providence, Rhode Island

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Prepared for: Picatinny Research Center
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FOREWORD

The present study is part of a program of ^{// *undupp*} "Theoretical Research on Combustion and Wave Propagation in Metal Dust-Oxidant Mixtures" being conducted by the Division of Engineering, Brown University, under United States Army Ordnance Contract No. DA-19-020-ORD-4761. The work was administered by the Pyrotechnics Laboratory of the Picattiny Arsenal, Dover, N. J.



SUMMARY

A macroscopic theory of two-phase flow with mass, momentum and energy exchange is discussed. The theory is applied to the study of systems which depart only slightly from local thermodynamic equilibrium. An example of wave propagation in a two-phase medium with viscous and thermal relaxation is calculated. It confirms some of the predictions of ref. 2, where a thermodynamic analysis was carried out.

1. Introduction The analysis presented here was undertaken in an attempt to understand better the nature of propagation of waves and combustion fronts in a pyrotechnic mixture. The mixture consists usually of two components, both in granular form: a light metal, such as aluminum or magnesium, and an oxidizer, such as potassium perchlorate. The granules have an average diameter of 15 microns which is usually much smaller than the smallest macroscopic scale of interest. (see ref. 1) For example, in a particular problem, the smallest macroscopic scale of interest, denoted by l , may be a fraction of a millimeter or larger. Furthermore, there are usually so many granules in a volume l^3 that the mixture may be considered macroscopically as a continuum. The implication of a continuum description of the granular mixture will be discussed more fully in the next section. For the moment we shall accept this view.

The mechanical and thermodynamic properties of such a continuum depend on the microscopic structure of the mixture; in particular, on the microscopic state of the aggregate of the granules. There are two extreme cases which may be discerned: In the first case, the mean distance between neighboring granules is large compared with the mean diameter of the granules; in the second, the two lengths are of the same order of magnitude. In the former case, the great majority of the granules are not in contact with another granule. These granules are merely particles suspended in a perfect gas. Such a mixture behaves essentially like a "Dusty Gas". In the second case, most of the granules are in direct contact with their neighboring granules. As an extreme example, the granules are so tightly packed together that they lock into one another. In this extreme case, the mixture behaves essentially like a solid, or better, a "porous solid" with the intergranular space filled with a perfect gas. In the intermediary range between the "dusty gas" and the "porous solid", the mixture assumes a mixed behavior. Thus, for example, the mixture exhibits

properties which are characteristic of a solid undergoing plastic deformation. The thermodynamics of a dusty gas and a porous solid are given in refs. 2 and 3; that of a loosely packed granular mixture is given in ref. 4.

In a thermodynamic analysis, it is assumed that the relative motion between the granules and the surrounding gas is negligible only. Furthermore, we assume that the granules and the surrounding gas are either in thermal equilibrium with one another at all times or are thermally insulated from each other. In spite of these rather severe restrictions, the result of a purely thermodynamic analysis often leads to important conclusions. An example of such an application is given in ref. 2.

In what follows we shall be interested in problems in which the granules and the gas between them are not in thermodynamic equilibrium. Between these two components, there will in general be an exchange in mass, momentum and energy. We propose to examine these non-equilibrium phenomena within the framework of a continuum theory.

Earlier contributions to this subject were mainly due to F. A. Williams who developed a statistical theory of spray and applied it to many combustion problems. (ref. 5, 6, 7) We adopt here an alternate approach -- the macroscopic approach. We shall make use of the method of irreversible thermodynamics to establish certain phenomenological laws describing the interaction between the flow and the particles suspended in it. The limitations of these laws are discussed. An application of the macroscopic theory to wave phenomena in such a medium is included.

2. Some Macroscopic Properties of the Mixture We shall now examine the conditions under which a granular mixture may be considered as a continuum on the macroscopic scale and we shall discuss some of the properties of such a continuum. This discussion can perhaps be best illustrated by considering a special instance.

Suppose that the average diameter of the granules is of the order of

10 microns, i.e., 10^{-3} cm, and the average distance between the neighboring granules is of the order of 100 microns, or 10^{-2} cm. It follows that in 1 cubic millimeter of the mixture there are about 1,000 granules. If the smallest macroscopic length of interest is greater than a half millimeter, to be abbreviated by $l (= 0.5 \text{ mm})$, then all linear dimensions equal to or less than l and all areas or volumes equal to or less than l^2 and l^3 respectively may be taken as macroscopically zero. Under such conditions, it may be possible to define the various macroscopic density variables at a point in the mixture considered as a continuum. Thus, for example, the mass density of the granules at a point may be defined as follows. Let δV be a macroscopic volume containing the point and δM be the total mass of the granules in δV . The ratio $\delta M/\delta V$ as $\delta V \rightarrow l^3$ (which is regarded as macroscopically zero) represents the mass density of the granules at a point of the mixture considered as a continuum. Likewise, the stress vector at a point in the mixture may be referred to the force acting on an element δA as $\delta A \rightarrow l^2$ etc. On the other hand, to define a mean temperature and velocity for the granules and the gas between the granules, we must exclude very rapid changes. An estimate of the smallest macroscopic time scale allowable for defining such quantities can be made as follows. First, let us consider a single granule of 10 micron diameter. Assuming a thermal diffusivity of $0.2 \text{ cm}^2/\text{sec}$ for the granule, it is seen that the thermal relaxation time in the granule is of the order of $(10^{-3})^2/0.2$ seconds or 5 microseconds. Hence if the smallest macroscopic time of interest is greater than 5 microseconds, we may assign a single temperature to each granule. Since all the granules in a volume l^3 undergo almost identical changes, we may in fact ascribe a mean temperature for all the granules in the volume. Similarly a mean velocity of the granules may be defined at each point of the mixture considered as a continuum. Next, let us examine the fluid surrounding the granules. Since the average distance between

neighboring granules is of the order of 100 microns or 10^{-2} cm. and the thermal diffusivity of gas is of the same order of magnitude as that in aluminum, namely, $0.2 \text{ cm}^2/\text{sec}$, therefore the thermal relaxation time in the gas between the granules is of the order of 5×10^{-4} sec. or half a millisecond. Hence, we may define a mean temperature for the gas surrounding the granules when the smallest macroscopic time of interest is at least 1 millisecond. Since the Prandtl number of the gas is of the order of unity, the viscous relaxation time is also about 5×10^{-4} seconds and we may ascribe a single velocity for the fluid inside a volume ρ^3 .

It follows from the above considerations that if the smallest macroscopic length of interest is large compared with the average distance between the granules and if the smallest macroscopic time of interest is long compared with the thermal relaxation time of the fluid between the granules then a macroscopic continuum description of the granular mixture is feasible.

There is one further point to be clarified. In the special example discussed, nothing is said about the time required for the granules and the gas surrounding them to attain the same temperature and velocity. We have only discussed the time required for defining a mean temperature and velocity for each of the two components of the mixture. An estimation of the time required for the granules and the surrounding gas to attain the same temperature can be readily made. First, we observe that the rate of heat transfer from the gas to a granule is of the order of

$$k_{\text{gas}} \frac{\Delta T}{D/2} \cdot \pi D^2 \quad (2.1)$$

where k_{gas} is the conductivity in the gas, ΔT is the temperature difference between the granules and the surrounding gas and D is the diameter of the granule. $\frac{\Delta T}{D/2}$ is thus a measure of the average temperature gradient at the surface of the granule. On the other hand, the total change in heat content in the granule as a result of heat transfer is

$$\left[\frac{4}{3} \pi \left(\frac{D}{2} \right)^3 \right] \rho \cdot C_p \Delta T \quad (2.2)$$

where the term in the bracket is clearly the volume of the granule while ρ and C_p are respectively the density and specific heat of the granule.

The time required for the establishment of thermal equilibrium between the two components is obtained by dividing (2.2) by (2.1), namely, $\frac{\pi}{3} \frac{\rho C_p}{k_{gas}} D^2$

If α denotes the thermal diffusivity in the granule ($\alpha = 0.2 \text{ cm}^2/\text{sec}$)

This time is $\frac{\pi}{3} \cdot \frac{k}{k_{gas}} \cdot \frac{D^2}{\alpha}$ where k is the conductivity of the granule.

For the case of an aluminum or magnesium granule, k is greater than k_{gas} by a factor of 1000. Since D^2/α is of the order of microseconds, the time required for the establishment of thermal equilibrium is about 5 milliseconds. By a similar analysis, we find the time required for the establishment of mechanical equilibrium is of the same order of magnitude.

The last calculation shows that if the smallest time scale of interest is greater than 10 milliseconds, the mixture can in fact be treated as a system in thermodynamic equilibrium. On the other hand, if the smallest time scale is of the order of 1 millisecond, we may define a mean temperature for the granule and another for the gas. The same applies to the velocity of the granule and the gas.

To be sure, we may visualize a situation in which the temperature and/or velocity in the gas surrounding the granules may vary considerably throughout most of the region enclosed by the small volume l^3 ; and, in spite of this, a mean temperature and velocity may be defined by some averaging process within the volume l^3 . If such is the case, the smallest macroscopic time of interest may be considerably smaller than a millisecond. However, the exact nature of the approximation used becomes rather unclear.

3. Conservation Laws The conservation laws governing the motion and deformation of the medium can be easily derived. For the sake of simplicity,

we shall consider the granules as all of one species and the gas as another. Generalization of the analysis to mixtures of many species does not present any special difficulties. We shall use the superscripts, the "primes" and "double-primes", to signify quantities pertaining to the granules and the surrounding gas respectively. Thus, ρ' and ρ'' denote respectively the mass of the granules and of the gas per unit volume of the mixture; \vec{u}' and \vec{u}'' denote respectively the velocities of the granules and of the gas.

The law of mass conservation can be expressed simply as:

$$\frac{\partial(\rho' + \rho'')}{\partial t} + \nabla \cdot (\rho' \vec{u}' + \rho'' \vec{u}'') = 0 \quad (3.1)$$

If \vec{f}' and \vec{f}'' denote respectively the body force acting on the granules and on the gas per unit volume of the mixture, the equation of momentum balance is

$$\frac{\partial}{\partial t} (\rho' \vec{u}' + \rho'' \vec{u}'') + \nabla \cdot (\rho' \vec{u}' \vec{u}' + \rho'' \vec{u}'' \vec{u}'') + \nabla p = \vec{f}' + \vec{f}'' \quad (3.2)$$

where p denotes the gas pressure.

In deriving the last equation, we have neglected a term which may be called the "pressure contributed by the granules". We recall that the quantity which is, on the macroscopic scale, called "pressure" is, in actuality, the average value of a component of the momentum flux on the microscopic scale. To be more precise, if we denote the velocity of a gas molecule relative to an observer moving with the mean velocity \vec{u}'' by \vec{c}'' and if m'' is the mass of the molecule, the pressure tensor p''_{ij} is defined by the phase average:

$$p''_{ij} = \langle m'' c''_i c''_j \rangle_{\vec{c}''}$$

where $\langle \rangle_{\vec{c}''}$ signifies an average over the velocity space. The scalar pressure p is by definition $\frac{1}{3} p''_{ii}$. In exactly the same manner, a pressure contributed by the random motion of the granules may be defined in terms of

$$p'_{ij} = \langle m' c'_i c'_j \rangle_{\vec{c}', m', T'}$$

where \vec{c}' is the velocity of a granule relative to an observer moving with the mean velocity \vec{u}' ; m' denotes the mass of the granules; and $\langle \rangle_{\vec{c}', m', T'}$ signifies an average taken over the phase space spanned by \vec{c}' , m' and the temperature of the granules T' . However, unlike the case of gas molecules \vec{c}' is usually small compared with \vec{u}' so that the "pressure" $p' = \frac{1}{3} \rho' \langle c'^2 \rangle$ is in general much smaller than $\rho' u'^2$. For this reason, we shall neglect p' the "pressure" due to the random motion of the granules.

Under the same approximation, we may write down the energy equation:

$$\begin{aligned} & \frac{\partial}{\partial t} \left[\rho' \left(U' + \frac{1}{2} u'^2 \right) + \rho'' \left(U'' + \frac{1}{2} u''^2 \right) \right] + \\ & + \nabla \cdot \left[\rho' \vec{u}' \left(U' + \frac{1}{2} u'^2 \right) + \rho'' \vec{u}'' \left(U'' + \frac{1}{2} u''^2 \right) \right] + \nabla \cdot (p \vec{u}'') = \\ & = \vec{f}' \cdot \vec{u}' + \vec{f}'' \cdot \vec{u}'' \end{aligned} \quad (3.3)$$

4. Equations Governing the Motion of the Granules and the Gas Let us define ω' and ω'' by the formulas:

$$\frac{\partial \rho'}{\partial t} + \nabla \cdot \rho' \vec{u}' = \omega' \quad (4.1)$$

$$\frac{\partial \rho''}{\partial t} + \nabla \cdot \rho'' \vec{u}'' = \omega'' \quad (4.2)$$

Evidently ω' has the significance of being the rate of increase of the total mass of the granules per unit volume of the mixture, while ω'' is the rate of increase of the mass of the gas per unit volume of the mixture. Eqs. (4.1) and (4.2) thus represent the equations of mass balance of the two components. Furthermore, by virtue of (3.1) that

$$\omega' + \omega'' = 0 \quad (4.3)$$

In a similar manner, we define \vec{F}' and \vec{F}'' by

$$\frac{\partial \rho' \vec{u}'}{\partial t} + \nabla \cdot (\rho' \vec{u}' \vec{u}') = \vec{f}' + \vec{F}' \quad (4.4)$$

$$\frac{\partial \rho'' \vec{u}''}{\partial t} + \nabla \cdot (\rho'' \vec{u}'' \vec{u}'') = -\nabla p + \vec{f}'' + \vec{F}'' \quad (4.5)$$

\vec{F}' is thus the total force exerted by the gas on the granules per unit volume of the mixture and \vec{F}'' is the reaction of this force on the gas. Evidently

$$\vec{F}' + \vec{F}'' = 0 \quad (4.6)$$

Furthermore, eqs. (4.4) and (4.5) may be interpreted as the equations of motion of the granules and of the gas respectively.

Finally, we define q' and q'' by the formulas:

$$\frac{\partial}{\partial t} \left[\rho' \left(U' + \frac{1}{2} u'^2 \right) \right] + \nabla \cdot \left[\rho' \vec{u}' \left(U' + \frac{1}{2} u'^2 \right) \right] = (\vec{f}' + \vec{F}') \cdot \vec{u}' + q' \quad (4.7)$$

$$\frac{\partial}{\partial t} \left[\rho'' \left(U'' + \frac{1}{2} u''^2 \right) \right] + \nabla \cdot \left[\rho'' \vec{u}'' \left(U'' + \frac{1}{2} u''^2 \right) \right] = -\nabla \cdot (p \vec{u}'') + (\vec{f}'' + \vec{F}'') \cdot \vec{u}'' + q'' \quad (4.8)$$

If we interpret (4.7) and (4.8) as the energy equations for the granules and the gas, the quantity q' will then have the significance of being the heat flow into the granules per unit volume of the mixture, while q'' is that flow into the gas per unit volume of the mixture. From (3.3), we see that

$$q' + q'' + \vec{F}' \cdot \vec{u}' + \vec{F}'' \cdot \vec{u}'' = 0 \quad (4.9)$$

Eqs. (4.3), (4.6), (4.9) provide three relations for the six variables: $\omega', \omega'', q', q'', \vec{F}', \vec{F}''$. We need three more relations before the problem is completely determined. In general, these relations must be postulated. However, when the state of the mixture departs only slightly from the state of local thermodynamic equilibrium, the form of those relations can be determined from thermodynamic considerations. (See the next section.)

We note that eq. (4.4) can also be written as:

$$\rho' \frac{d \vec{u}'}{dt'} + \omega' \vec{u}' = \vec{f}' + \vec{F}' \quad (4.10)$$

where $\frac{d}{dt'} = \frac{\partial}{\partial t} + \vec{u}' \cdot \nabla$. Likewise, if $\frac{d}{dt''} = \frac{\partial}{\partial t} + \vec{u}'' \cdot \nabla$ we

have:

$$\rho'' \frac{d\vec{u}''}{dt''} + \omega'' \vec{u}'' = -\nabla p + \vec{f}'' + \vec{F}'' \quad (4.11)$$

Similarly we can rewrite (4.7) and (4.8) as

$$\rho' \frac{d}{dt'} \left(U' + \frac{1}{2} u'^2 \right) + \omega' \left(U' + \frac{1}{2} u'^2 \right) = (\vec{f}' + \vec{F}') \cdot \vec{u}' + q' \quad (4.12)$$

$$\rho'' \frac{d}{dt''} \left(U'' + \frac{1}{2} u''^2 \right) + \omega'' \left(U'' + \frac{1}{2} u''^2 \right) = -\nabla \cdot (\rho \vec{u}'') + (\vec{f}'' + \vec{F}'') \cdot \vec{u}'' + q'' \quad (4.13)$$

Furthermore, if we eliminate $\vec{f}' + \vec{F}'$ from (4.10) and (4.12), we obtain

$$\rho' \frac{dU'}{dt'} + \omega' \left(U' - \frac{1}{2} u'^2 \right) = q' \quad (4.14)$$

Likewise, from (4.11) and (4.13)

$$\rho'' \frac{dU''}{dt''} + \omega'' \left(U'' - \frac{1}{2} u''^2 \right) = -p \nabla \cdot \vec{u}'' + q'' \quad (4.15)$$

The last equation may also be rewritten as

$$\rho'' \left[\frac{dU''}{dt''} + p \frac{d}{dt''} \left(\frac{1}{\rho''} \right) \right] + \omega'' \left(H'' - \frac{1}{2} u''^2 \right) = q'' \quad (4.16)$$

where use has been made of (4.2). In terms of the entropies, eqs. (4.14)

and (4.16) assume the simple form

$$\rho' \frac{dS'}{dt'} = \frac{1}{T'} \left[q' - \omega' \left(H' - \frac{1}{2} u'^2 \right) \right] \quad (4.17)$$

$$\rho'' \frac{dS''}{dt''} = \frac{1}{T''} \left[q'' - \omega'' \left(H'' - \frac{1}{2} u''^2 \right) \right] \quad (4.18)$$

since

$$T' dS' = dU' \quad , \quad U' = H' \quad (4.19)$$

for a rigid granule and $T'' dS'' = dU'' + p d\left(\frac{1}{\rho''}\right)$ for a gas.

5. Entropy Production

Let ρ be the density of the mixture. Then

$$\rho = \rho' + \rho''$$

It is convenient to introduce the mass fraction γ' which is defined as the total mass of the granules in a unit mass of the mixture. It follows that

$$\gamma' = \rho' / \rho \quad (5.2)$$

Likewise if γ'' denotes the total mass of gas in a unit mass of the mixture, we have

$$\gamma'' = \rho'' / \rho \quad (5.3)$$

Obviously,

$$\gamma' + \gamma'' = 1 \quad (5.4)$$

We shall also define the velocity \vec{u} of the mixture by the formula

$$\rho \vec{u} = \rho' \vec{u}' + \rho'' \vec{u}'' \quad (5.5)$$

Eq. (4.1) can then be rewritten as

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \rho \vec{u} = 0 \quad (5.6)$$

which is the continuity equation for the mixture. Finally, let us introduce

$$\frac{D}{Dt} = \frac{\partial}{\partial t} + \vec{u} \cdot \nabla \quad (5.7)$$

and

$$\vec{v}' = \vec{u}' - \vec{u}, \quad \vec{v}'' = \vec{u}'' - \vec{u} \quad (5.8)$$

It may be easily verified that (4.1) and (4.2) may be rewritten as:

$$\frac{D\gamma'}{Dt} + \frac{1}{\rho} \nabla \cdot (\rho \gamma' \vec{v}') = \omega' / \rho \quad (5.9)$$

$$\frac{D\gamma''}{Dt} + \frac{1}{\rho} \nabla \cdot (\rho \gamma'' \vec{v}'') = \omega'' / \rho \quad (5.10)$$

Also (4.18), (4.19) become:

$$\gamma' \frac{DS'}{Dt} + \gamma' \vec{v}' \cdot \nabla S' = \frac{1}{\rho T'} \left[q' - \omega' \left(H' - \frac{1}{2} u'^2 \right) \right] \quad (5.11)$$

$$\gamma'' \frac{DS''}{Dt} + \gamma'' \vec{v}'' \cdot \nabla S'' = \frac{1}{\rho T''} \left[q'' - \omega'' \left(H'' - \frac{1}{2} u''^2 \right) \right] \quad (5.12)$$

If S denotes the entropy of a unit mass of the mixture,

$$S = \gamma' S' + \gamma'' S'' \quad (5.13)$$

and we may compute $\frac{DS}{Dt}$ from (5.9), (5.10), (5.11), (5.12) and (5.13).

Omitting the algebraic details, we find ultimately the equation

$$\rho \frac{DS}{Dt} + \nabla \cdot (\rho' \vec{v}' S' + \rho'' \vec{v}'' S'') = \Phi \quad (5.14)$$

where

$$\Phi = \frac{q'}{T'} + \frac{q''}{T''} + \frac{\omega'}{T'} \left(-G' + \frac{1}{2} u'^2 \right) + \frac{\omega''}{T''} \left(-G'' + \frac{1}{2} u''^2 \right) \quad (5.15)$$

and G' , G'' are Gibbs' free energy per unit mass of the granules and of the gas respectively. Eq. (5.14) has a simple meaning, for it may be rewritten as

$$\frac{\partial \rho S}{\partial t} + \nabla \cdot (\rho' \vec{u}' S' + \rho'' \vec{u}'' S'') = \Phi \quad (5.16)$$

where use has been made of (5.6) and (5.8). If we integrate (5.16) over a region enclosed by a smooth control surface, we have

$$\frac{\partial}{\partial t} \int S \rho d\tau + \int (\rho' \vec{u}' S' + \rho'' \vec{u}'' S'') \cdot d\vec{A} = \int \Phi d\tau \quad (5.17)$$

The first term of this equation is clearly the rate of increase of entropy inside the volume. The second term represents the amount of entropy convected out of the region. The last term must then be the entropy produced in the region. Consequently, Φ signifies the entropy produced in a unit volume of the mixture. Making use of (4.3), (4.6) and (4.9), we have:

$$\Phi = \frac{\vec{F}' \cdot (\vec{u}'' - \vec{u}')}{T''} + q' \left(\frac{1}{T'} - \frac{1}{T''} \right) + \omega' \left[\frac{G'' - \frac{1}{2} u''^2}{T''} - \frac{G' - \frac{1}{2} u'^2}{T'} \right] \quad (5.18)$$

6. Small Departure from Thermodynamic Equilibrium At thermodynamic equilibrium,

$$\begin{aligned} \vec{u}' &= \vec{u}'' & , \vec{F}' &= \vec{F}'' = 0 \\ T' &= T'' & , q' &= q'' = 0 \\ G' &= G'' & , \omega' &= \omega'' = 0 \end{aligned} \quad (6.1)$$

so that $\Phi = 0$ and the entropy of the system attains a maximum. When the system is near equilibrium, we can expect the following linear relation between the "fluxes" and the "forces".

$$\vec{F}' = -\vec{F}'' = k(\vec{u}'' - \vec{u}') \quad (6.2)$$

$$q' = -q'' = \lambda'(T'' - T') + \nu \left[\frac{G'' - \frac{1}{2} u''^2}{T''} - \frac{G' - \frac{1}{2} u'^2}{T'} \right] \quad (6.3)$$

$$\omega' = -\omega'' = \mu \left[\frac{G'' - \frac{1}{2} u''^2}{T''} - \frac{G' - \frac{1}{2} u'^2}{T'} \right] + \frac{\nu}{T''} [T'' - T'] \quad (6.4)$$

where $\lambda' > 0$, $\mu > 0$, $k > 0$ and $\lambda = \lambda' - \frac{\nu^2}{\mu T''} > 0$. In particular, if there is no reaction between the granules and the surrounding gas, we have simply:

$$\begin{aligned} F' &= -F'' = k(\vec{u}'' - \vec{u}') \\ q' &= -q'' = \lambda(T'' - T') \end{aligned} \quad (6.5)$$

On the other hand, if the smallest macroscopic scale is large compared with the time required for the granules and their surrounding gas to attain mechanical and thermal equilibrium, then $\vec{u}' = \vec{u}''$ and $T' = T''$. For this case, if there is mass exchange between the granules and the gas surrounding them, we have

$$\omega' = -\omega'' = \frac{\mu}{T''} (G'' - G') \quad (6.6)$$

Of course, in nearly all cases where chemical reaction enters as an important factor, $G'' - G'$ is not small and the system undergoes changes

which are very far from thermodynamic equilibrium. In such cases (6.6) cannot be used and must be replaced by an appropriate rate law determined from other considerations. On the other hand, the relaxation times between the granules and the surrounding gas are in general small and the state of the system is never too far from the state of thermodynamic equilibrium. This means that (6.2) and (6.3) may be used in most practical applications.

7. Characteristic Theory The basic equations governing the one-dimensional motion of the mixture are summarized below:

$$\frac{\partial \rho'}{\partial t} + \frac{\partial \rho' u'}{\partial x} = \omega' \quad (7.1)$$

$$\frac{\partial \rho''}{\partial t} + \frac{\partial \rho'' u''}{\partial x} = \omega'' \quad (7.2)$$

$$\frac{\partial \rho' u'}{\partial t} + \frac{\partial \rho' u'^2}{\partial x} = f' + F' \quad (7.3)$$

$$\frac{\partial \rho'' u''}{\partial t} + \frac{\partial \rho'' u''^2}{\partial x} = -\frac{\partial p}{\partial x} + f'' + F'' \quad (7.4)$$

$$\frac{\partial S'}{\partial t} + u' \frac{\partial S'}{\partial x} = \Phi' / \rho' \quad (7.5)$$

$$\frac{\partial S''}{\partial t} + u'' \frac{\partial S''}{\partial x} = \Phi'' / \rho'' \quad (7.6)$$

where

$$\Phi' = \frac{1}{T'} \left[q' - \omega' \left(H' - \frac{1}{2} u'^2 \right) \right] \quad (7.7)$$

$$\Phi'' = \frac{1}{T''} \left[q'' - \omega'' \left(H'' - \frac{1}{2} u''^2 \right) \right] \quad (7.8)$$

In addition, we have the equation of state. This can be readily shown to be

$$p'' = \frac{p}{RT''} \quad (7.9)$$

where R is the gas constant and T is the temperature of the gas. Furthermore, the entropies of the granules and the gas are respectively

$$S' = S'_0 + C'_p \log\left(\frac{T'}{T_0}\right) \quad (7.10)$$

$$S'' = S''_0 - C''_p \log\left(\frac{P}{P_0}\right)^{\frac{\gamma-1}{\gamma}} \left(\frac{T_0}{T''}\right) \quad (7.11)$$

(see ref. 2) To calculate the characteristics of the system, we shall denote the change of any variable along a characteristics by $\frac{\delta}{\delta t}$. Thus

$$\frac{\delta}{\delta t} = \frac{\partial}{\partial t} + c \frac{\partial}{\partial x} \quad (7.12)$$

where c denotes the speed of propagation of the characteristics surface. Let us now eliminate the spatial derivatives from all of the equations and look for the specific directions along which the variation of the different quantities can be determined by an equation in this direction. These are the characteristics directions. Thus, from (7.5), we find the characteristics direction in the $x-t$ -space is specified by the wave speed $c = u'$ and along which $\frac{\delta S'}{\delta t} = \Phi' / \rho'$. Carrying out this procedure, we obtain the following results. There are six sets of characteristics:-

- 1) A triplet of the characteristics $c = u'$ along which

$$\frac{\delta S'}{\delta t} = \Phi' / \rho' \quad (7.13)$$

and

$$\rho' \frac{\delta u'}{\delta t} = f' + F' - u' \omega' \quad (\text{double}) \quad (7.14)$$

- 2) A single family of characteristics propagating at $c = u''$ along which

$$\frac{\delta S''}{\delta t} = \Phi'' / \rho'' \quad (7.15)$$

- 3) Two families of characteristics propagating with the speeds $c = u'' \pm \sqrt{\frac{\delta P}{\rho''}}$ along which we have

$$\begin{aligned} & \rho'' \left[f'' + F'' + \omega'' (c - 2u'') \right] - \frac{\delta P}{\delta t} - c \frac{\delta \rho'' u''}{\delta t} + u''^2 \frac{\delta \rho''}{\delta t} + \\ & + \frac{\delta P}{c_p'' (c - u'')} \left[c \frac{\delta \Phi''}{\delta t} - u'' \frac{\delta S''}{\delta t} \right] = 0 \end{aligned} \quad (7.16)$$

Because of the degeneracy of the characteristics relations (7.14), we have only five equations for the six variables: S', S'', u', u'', p' and p''

Note that the pressure can always be calculated from the equation of state and expressed in terms of these variables.

To complete the system we need an additional equation. This is provided by (7.1) if we eliminate $\frac{\partial}{\partial t}$ from it with the help of (7.12). Thus we find that along $c = u'$

$$\frac{\delta p'}{\delta t} + p' \frac{\partial u'}{\partial x} = \omega' \quad (7.17)$$

These relations are adequate for studying wave problems in the mixture by numerical methods.

8. Small Amplitude Waves For small amplitude waves we may linearize the governing equations and an analytical solution may be constructed. In this calculation, we shall assume $\omega' = \omega'' = 0$. The linearized form of the governing equations are:

$$\frac{\partial p'}{\partial t} + p'_0 \nabla \cdot u' = 0 \quad (8.1)$$

$$\frac{\partial p''}{\partial t} + p''_0 \nabla \cdot u'' = 0 \quad (8.2)$$

$$p'_0 \frac{\partial u'}{\partial t} + p''_0 \frac{\partial u''}{\partial t} = -\nabla p \quad (8.3)$$

$$p'_0 \frac{\partial u'}{\partial t} = k(\vec{u}'' - \vec{u}') \quad (8.4)$$

$$p'_0 c'_p \frac{\partial T'}{\partial t} + p''_0 c''_p \frac{\partial T''}{\partial t} = \frac{\partial p}{\partial t} \quad (8.5)$$

$$p'_0 c'_p \frac{\partial T'}{\partial t} = \lambda(T'' - T') \quad (8.6)$$

$$\frac{p''}{p_0} = \frac{p''}{p''_0} + \frac{T''}{T''_0} \quad (8.7)$$

where subscript "0" characterizes the undisturbed field. We have assumed that the body forces: $\vec{f}' = \vec{f}'' = 0$. In

this section $p, \vec{u}', \vec{u}'', \vec{p}', \vec{p}'', T', T''$

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to be the perturbed quantities.

If \vec{u}' , T' , ρ'' and T'' are eliminated from the above system of equations, we have

$$\left(\frac{\partial}{\partial t} + \zeta\right) \frac{\partial}{\partial t} \left(\frac{P}{\gamma P_0}\right) = -\left(\frac{\partial}{\partial t} + \zeta\right) \nabla \cdot \vec{u} \quad (8.8)$$

$$a_0^2 \left(\frac{\partial}{\partial t} + \beta\right) \nabla \left(\frac{P}{\gamma P_0}\right) = -\left(\frac{\partial}{\partial t} + \alpha\right) \frac{\partial \vec{u}}{\partial t} \quad (8.9)$$

where

$$\alpha = k \left(\frac{1}{\rho'_0} + \frac{1}{\rho''_0} \right) \quad (8.10)$$

$$\beta = k / \rho'_0 \quad (8.11)$$

$$\zeta = \lambda \left(\frac{1}{\rho''_0 c''_v} + \frac{1}{\rho'_0 c'_p} \right) \quad (8.12)$$

$$\zeta_1 = \lambda \left(\frac{1}{\rho''_0 c''_p} + \frac{1}{\rho'_0 c'_p} \right) \quad (8.13)$$

Eliminating \vec{u} we obtain

$$\left[\frac{\partial^2}{\partial t^2} \boxed{a} + \left(\frac{1}{\tau_1} + \frac{1}{\tau_2} \right) \frac{\partial}{\partial t} \boxed{a} + \frac{1}{\tau_1 \tau_2} \boxed{a} \right] \frac{P}{\gamma P_0} = 0 \quad (8.14)$$

where

$$\boxed{a} = \frac{\partial^2}{\partial t^2} - a^2 \nabla \quad (8.15)$$

$$\tau_1 = 1/\alpha \quad (8.16)$$

$$\tau_2 = 1/\zeta \quad (8.17)$$

$$a_1 = a_0 \sqrt{\frac{\beta + \zeta_1}{\alpha + \zeta}} \quad (8.18)$$

$$a_2 = a_0 \sqrt{\frac{\beta \zeta_1}{\alpha \zeta}} \quad (8.19)$$

and $a_0 = \sqrt{\gamma P_0 / \rho''_0}$ is the sound speed in the gas when there is no granules.

The solution of (8.14), satisfying the initial condition that the medium is undisturbed at $t=0$ and the boundary condition associated with the

piston problem, i.e.,

$$u' = u_0 H(t) \quad (8.20)$$

at $x=0$, is

$$p = \frac{\rho_0'' u_0}{2\pi i} \int_{-i\infty}^{i\infty} \frac{1}{\Lambda} \frac{\alpha+s}{\beta+s} e^{st-\Lambda x} ds \quad (8.21)$$

where

$$\Lambda = \frac{s}{a_0} \sqrt{\frac{(s+\alpha)(s+\zeta)}{(s+\beta)(s+\zeta_1)}} \quad (8.22)$$

For large s ,

$$st - \Lambda x = s\left(t - \frac{x}{a_0}\right) - \frac{(\alpha-\beta) + (\zeta-\zeta_1)}{2} \frac{x}{a_0} + O\left(\frac{1}{s}\right) \quad (8.23)$$

Hence, if $x > a_0 t$ the integral must be closed to the right of the imaginary axis and hence, $p=0$. For $x < a_0 t$, the integral must be closed to the left and p is, in general, different from zero. In fact, the value of p as $x \rightarrow a_0 t - 0$ is easily computed by applying a Tauberian theorem.

Thus, it is found that

$$\begin{aligned} \lim_{x \rightarrow a_0 t - 0} \frac{p}{\gamma \rho_0} &= \frac{u_0}{a_0} \exp\left[-\frac{(\alpha-\beta) + (\zeta-\zeta_1)}{2} t\right] \\ &= \frac{u_0}{a_0} \exp\left[-\frac{1}{2} \left(\frac{k}{\rho_0''} + \frac{\gamma-1}{\gamma} \frac{\lambda}{\rho_0'' c_0}\right) t\right] \end{aligned} \quad (8.24)$$

Hence the wave front is rapidly damped. This is as it should be; for, the wave front represents a signal propagating between the granules. The probability that it is not scattered by the granules decreases exponentially with time.

To examine the behavior of p at the piston for large t , we introduce a new variable $s' = ts$. Thus, at the piston,

$$p = \frac{\rho_0'' u_0 a_0}{2\pi i} \int_{-i\infty}^{i\infty} \frac{\sqrt{\frac{s'+\alpha t}{s'+\beta t} \frac{s'+\zeta t}{s'+\zeta_1 t}}}{s'} e^{s' \frac{ds'}{s'}} \quad (8.25)$$

so that as $t \rightarrow \infty$

$$\lim_{t \rightarrow \infty} p_{\text{piston}} = \rho_0'' u_0 a_0 \sqrt{\frac{\alpha}{\beta} \frac{\zeta_1}{\zeta}} \quad (8.26)$$

Hence

$$\lim_{t \rightarrow \infty} \left(\frac{P_{\text{piston}}}{\gamma_{\text{Mix}} P_0} \right) = \frac{U_0}{a_2} \quad (8.27)$$

where

$$\gamma_{\text{Mix}} = \gamma \frac{\zeta_1}{\zeta_2} \quad (8.28)$$

The last formula shows that for large time the pressure level is determined by the equilibrium sound speed a_2 . On the other hand, for small time the pressure at the piston is determined by

$$\frac{P_{\text{piston}}}{\gamma P_0} = \frac{U_0}{a_0} \left[1 - \frac{(\alpha - \beta) + (\zeta_2 - \zeta_1)}{2} t + O(t^2) \right] \quad (8.29)$$

Finally, the wave profile at large time can also be deduced by using the method of the steepest descent. (For details, see ref. 5).

Let us introduce the function

$$W(s) = s \left[1 - \frac{x}{a_0 t} \sqrt{\frac{s + \alpha}{s + \beta} \frac{s + \zeta_1}{s + \zeta_2}} \right] \quad (8.30)$$

then

$$p = \frac{P_0 U_0}{2\pi i} \int_{-i\infty}^{i\infty} \frac{1}{\Lambda} \frac{\alpha + s}{\beta + s} e^{t W(s)} ds \quad (8.31)$$

As $t \rightarrow \infty$, the main part of the contribution of the integrand is concentrated near the point where $W(s)$ is a maximum. Since W is an analytic function of s , this point is in actuality a saddle point. A careful examination of the trajectory of the relevant saddle point shows that it is located on the real axis and crosses $s = 0$ at

$$\frac{x}{a_0 t} = \sqrt{\frac{\beta \zeta_1}{\alpha \zeta_2}} = \frac{a_2}{a_0} \quad (8.32)$$

Now the point $s = 0$ is a pole of the integrand, it follows that, for $x < a_2 t$ the solution is of one type while, for $x > a_2 t$, the solution is of a different type. Taking into account the confluence of the saddle point and the simple pole at $s = 0$, we find

$$\frac{p}{\gamma_{\text{Mix}} p_0} = \frac{1}{2} \frac{u_0}{a_2} \left\{ e^{w_0 t} \frac{|s_0|}{s_0} e^{\frac{1}{2} w_0'' s_0^2 t} \operatorname{erfc} \left(\sqrt{\frac{1}{2} w_0'' s_0^2 t} \right) + 2H \left(1 - \frac{x}{a_2 t} \right) \right\} \quad (8.33)$$

where s_0 is the saddle point and $w_0 = W(s_0)$. Since $s_0 > 0$ for $x < a_0 t$ and $s_0 < 0$ for $x > a_0 t$, the wave profile consists of a small precursor wave propagating at the sound speed a_0 which is followed by the main signal propagating at the equilibrium speed a_2 . This conclusion is qualitatively similar to the result of G. F. Carrier (ref. 8) who considered the same problem without thermal relaxation. The main difference is that the main signal propagates with the equilibrium sound speed a_2 here. We note that the same conclusion was deduced earlier in ref. 2 where a Thermodynamic analysis was made.

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