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COMPOSITE SPECIES IN THE HYDRODYNAMIC THEORY OF ATOMIC
MIXING IN MULTICOMPONENT PARTIALLY IONIZED GASES

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ABSTRACT

A dynamical description of atomic mixing in multicomponent gases and plasmas was summarized in a previous report (UCRL-ID-145502). That description is based on the use of separate continuity and momentum equations for each species present, including neutral atoms, ions, and free electrons. This level of detail is not always feasible in practical problems, where subsets of species (e.g., neutral atoms of a particular element together with their ionization products) must be grouped or lumped together into composite species or materials to make the problem tractable. A simple procedure for constructing such composite species was outlined in UCRL-ID-145502, but not in sufficient detail to enable implementation. In particular, the treatment of the free electrons presents some subtleties, since they cannot be included in the composite species for dynamical purposes, whereas they are ordinarily lumped together with the atoms and ions that produced them for state equation purposes. Our purpose here is to provide a more complete description of the procedure by which composite species and their evolution equations may be defined and derived. Special attention is given to the problem of how to deal with the free electrons in a manner consistent with the different roles they play in the dynamics and thermodynamics. The results are given in a form that should be well suited for implementation in hydrodynamics codes that use conventional material state equations.

1. INTRODUCTION

The dynamics of multicomponent gases and plasmas is usually described in terms of momentum and energy transport equations for the fluid mixture as a whole, continuity equations for each species or component in the mixture, and constitutive relations for the fluxes of momentum, energy, and species masses due to molecular collisions. These constitutive relations are ordinarily diffusional in character. For such a description to be valid, the collisional transfer of momentum and/or energy between the different species must be fast relative to time scales of interest. When this condition is not satisfied, it becomes necessary to use a more general description in which each species has its own momentum and/or energy transport equation. The conventional diffusional description of species transport is simply an approximation to these individual species momentum equations [1,2].

In order to simulate the atomic mixing of materials in fast processes where the diffusion approximation is not valid, it is necessary to solve the individual species momentum equations. These equations were summarized in a previous report [3]. In the present context, each type of ion and neutral atom, as well as the free electrons, must be considered a separate species. Thus, for example, even a pure argon plasma is actually a multicomponent mixture consisting of several different species, namely neutral Ar atoms, singly, doubly, etc. ionized argon ions (Ar^+ , Ar^{++} , etc.), and free electrons (e^-). We shall denote the various types of atoms present by the symbol X_k ($k = 1, 2, \dots$), and the various ionization states of X_k by X_k^n ($n = 0, 1, 2, \dots$), so that X_k^0 refers to neutral atoms of X_k while X_k^n for $n \geq 1$ refers to n -tuply ionized X_k ; i.e., $X_k^1 = X_k^+$, $X_k^2 = X_k^{++}$, etc. The free electrons will simply be denoted by the symbol e , which will also be used as a sub- or superscript.

It is clear that a plasma produced by ionizing a mixture of many different types of atoms X_k will contain a much larger number of different species X_k^n and e . Due to computer time and storage constraints, it is not always feasible in practical calculations to separately track each and every one of these species. The usual practice has been to simplify the description by assuming ionization equilibrium and considering each type of neutral atom X_k^0 , together with all of its ionization states or products X_k^n ($n = 1, 2, \dots$) and the free electrons thereby produced, as a single composite species or material X_k (which will be referred to for short as composite species or material k). The mixture as a whole may then be regarded as a mixture of a much smaller number of these various composite species. Thus, for example, a partially ionized mixture of argon and helium would be considered a mixture of the two composite species Ar and He, while a pure argon plasma would consist of the single composite species Ar and hence would no longer be formally regarded as a multicomponent mixture.

By construction, each composite species X_k is inherently electrically neutral, and therein lies a dilemma. As is well known, an inhomogeneous electrically neutral plasma will spontaneously develop a nonzero internal electric field \mathbf{E} , which serves as the mechanism by which charge neutrality is enforced [2–4]. The resulting electric field exerts forces on the charged species. Those forces appear in the momentum equations, and they significantly affect the motion of the charged species and hence the dynamics of the plasma as a whole. Within the context of a diffusional description, this effect is usually referred to as “ambipolar diffusion” [4]. The dilemma is that since the composite species X_k are electrically neutral, they cannot and do not interact with the electric field. If the plasma dynamics were described entirely in terms of the composite species X_k , the effects of the electric forces would therefore be entirely lost, and this would represent an inaccurate and unacceptable approximation.

In order to retain the electric forces, it is clear that the free electrons cannot be lumped together with the ions that produced them for dynamical purposes; i.e., in the momentum equations. In contrast, however, thermodynamic state relations for the composite species X_k ordinarily *do* include the effects of the free electrons. This dichotomy is inconvenient and a bit cumbersome, but it is not fatal and can be dealt with by carefully transforming back and forth, as necessary, between the various quantities that do and do not include the contributions of the free electrons. Our main purpose here is to explicitly show how this may be done, and to present the resulting relations for doing so in sufficient detail to permit their incorporation into hydrodynamics codes.

The present discussion is therefore a sequel to Ref. [3], with which the reader is presumed to be familiar. Unless otherwise stated, the notation is essentially the same as that of Ref. [3], and all equations are again written in cgs units. We also take this opportunity to remark parenthetically that the relations given in Ref. [3] for the various coupling coefficients, which will also be employed here, were derived under the assumption that the relative velocities between different species are not too large. When this assumption is not valid, the relations in Ref. [3] are no longer accurate and should be replaced by the corrected relations given by Burgers [5] and Chang [6].

2. THERMODYNAMIC STATE RELATIONS

Thermodynamic state relations for each composite species or material by itself, unmixed with other composite species, may be determined by a conventional thermodynamic equilibrium calculation in which the appropriate free energy or thermodynamic potential of

material X_k is minimized with respect to the relative populations of the neutral atoms, ions, and free electrons of which it is composed, subject to the constraints that its nuclei and electrons are conserved during ionization and recombination. Thus each “pure” composite species X_k has a thermal equation of state for its pressure $\tilde{p}_k(\tilde{\rho}_k, T)$ as a function of its mass density $\tilde{\rho}_k$ and temperature T , a caloric equation of state for its specific internal energy $\tilde{e}_k(\tilde{\rho}_k, T)$, and so on. The tilde \sim is used to indicate quantities associated with a “pure” composite material, unmixed with other such materials. These quantities of course differ from the corresponding *partial* quantities (e.g., the partial pressure p_k and partial density ρ_k) that pertain to composite material X_k within a multimaterial mixture.

The aforementioned equilibrium calculation of course also determines the number densities \tilde{n}_k^n of the ionization states X_k^n present within material X_k , or equivalently the corresponding mass densities $\tilde{\rho}_k^n = m_k^n \tilde{n}_k^n$, where m_k^n is the mass of a single particle of X_k^n . Clearly $m_k^n = m_k^0 - nm_e$, where m_k^0 is the mass of a single neutral atom of X_k^0 and m_e is the mass of a single electron. Since the ionization states X_k^n can be numerous, it would be cumbersome for standardized state routines or packages to provide all of the number densities \tilde{n}_k^n , especially since they are not required in most applications. These quantities are therefore not usually accessible to the user. However, the mean degree of ionization is normally available, and is indeed required for the development that follows. This quantity is usually expressed in terms of either the electron number density $\tilde{n}_k^e(\tilde{\rho}_k, T)$, or the mean positive charge per heavy particle $|Q_e|Z_k^*(\tilde{\rho}_k, T)$, where $Z_k^* = \sum_n n \tilde{n}_k^n / \tilde{n}_k^h$, $\tilde{n}_k^h = \sum_n \tilde{n}_k^n$ is the total number density of heavy particles in material k , and $Q_e < 0$ is the charge on a single electron. It is easy to see that the two quantities \tilde{n}_k^e and Z_k^* contain equivalent information. Each particle of X_k^n gives rise to n free electrons, so that $\tilde{n}_k^e = \sum_n n \tilde{n}_k^n$ and hence $Z_k^* = \tilde{n}_k^e / \tilde{n}_k^h$.

Moreover, the number of heavy particles is not changed by ionization, so \tilde{n}_k^h has the same value that it would have in the absence of ionization, namely $\tilde{n}_k^h = \tilde{\rho}_k/m_k^0$. It follows that

$$\tilde{n}_k^e = \frac{Z_k^* \tilde{\rho}_k}{m_k^0} \quad (1)$$

which is the simple basic relation between \tilde{n}_k^e and Z_k^* . In what follows, we shall assume the availability of either $\tilde{n}_k^e(\tilde{\rho}_k, T)$ or $Z_k^*(\tilde{\rho}_k, T)$, from which the other may easily be obtained from Eq. (1).

Although ionization does not change the number of heavy particles, it does slightly change their masses. The mean mass of a heavy particle of material k is also simply related to Z_k^* , and is given by $m_k^h = \sum_n m_k^n \tilde{n}_k^n / \tilde{n}_k^h = \sum_n (m_k^0 - n m_e) \tilde{n}_k^n / \tilde{n}_k^h = m_k^0 - m_e \tilde{n}_k^e / \tilde{n}_k^h = m_k^0 - Z_k^* m_e$. Moreover, the mass densities of free electrons and heavy particles within material k are clearly given by $\tilde{\rho}_k^e = m_e \tilde{n}_k^e$ and $\tilde{\rho}_k^h = \tilde{\rho}_k - \tilde{\rho}_k^e$, respectively, from which it follows that $m_k^h \tilde{n}_k^h = m_k^0 \tilde{n}_k^h - m_e \tilde{n}_k^e = \tilde{\rho}_k^h$.

As discussed above, the free electrons cannot be lumped together with the heavy particles in the momentum equations, which describe the motion of the heavy particles only. Correspondingly, the pressure forces in the momentum equations must be evaluated with the electronic contributions excluded. It is therefore necessary to separate out the electronic contributions to the functions \tilde{p}_k , which in turn requires an additivity assumption; i.e., $\tilde{p}_k = \tilde{p}_k^h + \tilde{p}_k^e$. Thus we must also assume that the state function $\tilde{p}_k^e(\tilde{\rho}_k, T)$ for the electron pressure of material k is known (albeit perhaps only approximately). The heavy-particle pressures are then simply given by $\tilde{p}_k^h = \tilde{p}_k - \tilde{p}_k^e$. It should be noted, however, that \tilde{p}_k^h is not yet the pressure whose gradient appears in the momentum equation for material k . The

pressure forces in gas mixtures are proportional to gradients of *partial* pressures [3], which have not yet been defined.

Up to this point, we have tacitly assumed that the electrons and heavy particles have the same common temperature T . When this is not the case the situation becomes somewhat more complicated, but the basic ideas remain the same. For present purposes, the only essential difference is that the various thermodynamic quantities become functions of both the heavy-particle temperature T and the electron temperature T_e .

The preceding discussion pertains to the state relations for “pure” composite materials in isolation, and we shall assume that these relations are available for all such materials present, either in analytical or tabular form. In the present context, however, we are concerned with mixtures of several such materials, for which we require similar thermodynamic state relations. Strictly speaking, the state relations of a multicomponent mixture cannot in general be expressed in terms of the state relations of the pure materials of which it is composed. (The reason is simply that the state relations of a mixture depend upon the form of the interatomic forces between particles of different materials, and this information does not enter into, and hence cannot be extracted from, the state relations of the pure materials.) Unfortunately, it is rarely feasible to construct accurate state relations for multicomponent mixtures of interacting components, and even less feasible to employ them in practical hydrodynamic calculations. In lieu of this, one must perforce approximate the state relations of the mixture in terms of those of the pure materials of which it is composed. The question of how best to do so is nontrivial, and will be discussed in detail elsewhere [7]. For present purposes, it suffices to note that such approximations are ordinarily based on an artificial partitioning or separation of the mixture into its constituent materials, in which material k

is regarded as being confined by itself within a compartment or subvolume with volume fraction α_k . Various approximate procedures have been proposed for determining these volume fractions, but they need not concern us here. We simply regard the α_k as known quantities, the actual values of which are immaterial for present purposes.

In terms of its volume fraction, the partial mass density of composite material k in the mixture is related to its corresponding pure material mass density by

$$\rho_k = \alpha_k \tilde{\rho}_k \quad (2)$$

and the total mass density of the mixture is simply $\rho = \sum_k \rho_k$. The partial mass density of material k exclusive of its free electrons is given by

$$\rho_k^h = \alpha_k \tilde{\rho}_k^h = \rho_k - \rho_k^e \quad (3)$$

where $\rho_k^e = \alpha_k \tilde{\rho}_k^e = m_e n_k^e$ is the partial mass density of the electrons associated with composite species k , and $n_k^e = \alpha_k \tilde{n}_k^e$ is the corresponding partial number density. Similarly, the partial number density of the heavy particles of material k is simply $n_k^h = \alpha_k \tilde{n}_k^h$. The total mass density of the free electrons in the mixture is then simply $\rho_e = \sum_k \rho_k^e = m_e n_e$, where $n_e = \sum_k n_k^e$ is the total number density of the free electrons.

Regardless of how the α_k are defined, the total pressure of the mixture is invariably taken to be

$$p = \sum_k \alpha_k \tilde{p}_k \quad (4)$$

a relation for which there is in fact a sound theoretical basis [7]. The quantities $\alpha_k \tilde{p}_k$ therefore

play the role of partial pressures, since their sum is the total pressure. Moreover, it is easy to verify that $\alpha_k \tilde{p}_k$ reduces to the usual partial pressure of material k in a mixture of ideal gases, where $\tilde{p}_k = \tilde{n}_k^h k_B T + \tilde{n}_k^e k_B T_e$, and k_B is Boltzmann's constant. The partial pressure of material k in the mixture will therefore be taken to be

$$p_k = \alpha_k \tilde{p}_k \tag{5}$$

The partial pressure of material k *sans* electrons is then given by

$$p_k^h = \alpha_k (\tilde{p}_k - \tilde{p}_k^e) = \alpha_k \tilde{p}_k^h \tag{6}$$

which now indeed is the pressure whose gradient appears in the momentum equation for material k *sans* electrons (see Sect. 4 below).

3. CONTINUITY EQUATIONS

In general, the continuity equations in a multicomponent mixture contain source terms representing the exchange of mass between different species due to chemical reactions, including ionization and recombination. In Ref. [3], these chemical source terms were omitted for simplicity, but they must be carefully and correctly accounted for in real calculations. There is a common misconception, occasionally found even in textbooks, that these source terms vanish in chemical equilibrium, but this is not in general the case. What actually happens is that the kinetic expressions for these source terms assume an indeterminate form in the equilibrium limit, while the source terms themselves remain nonzero and are implicitly

determined by the equilibrium constraint conditions [8]. These conditions are expressed by the law of mass action, or in the present context by Saha equations.

The chemical source terms in the continuity equations are constrained by the fact that mass is conserved in chemical reactions. This in turn implies that these source terms must add up to zero when they are summed over any subset of species which is chemically decoupled from all other species outside the subset. Each composite species X_k constitutes just such a subset, and hence its partial density ρ_k satisfies a continuity equation in which no chemical source terms appear. In contrast, the partial densities ρ_k^h do not include the free electrons, and their continuity equations consequently contain source terms that represent the interconversion between free and bound electrons by ionization/recombination processes. Of course, since $m_e \ll m_k^0$ these source terms are very small, but they may as well be completely eliminated by solving continuity equations for the partial densities ρ_k rather than ρ_k^h ; i.e.,

$$\frac{\partial \rho_k}{\partial t} + \nabla \cdot (\rho_k \mathbf{u}_k) = 0 \quad (7)$$

where \mathbf{u}_k is the mean velocity of composite material k *including* its free electrons.

It is conceptually essential to note that \mathbf{u}_k differs slightly from the velocity of material k *sans* electrons, which is denoted by \mathbf{u}_k^h . It is the velocities \mathbf{u}_k^h , rather than \mathbf{u}_k , that will be determined by the momentum equations. The difference between \mathbf{u}_k and \mathbf{u}_k^h is small because m_e is small, but there is no need to neglect it since the relation between the two velocities can easily be calculated exactly. The momentum density of composite species k including its free electrons is given by

$$\rho_k \mathbf{u}_k = \rho_k^h \mathbf{u}_k^h + \rho_k^e \mathbf{u}_e \quad (8)$$

where \mathbf{u}_e is the mean velocity of the electrons, which of course is independent of k (the free electrons are indistinguishable) and is determined by Eq. (10) of Ref. [3]. In the present context and notation, this relation becomes

$$q_e \rho_e \mathbf{u}_e = - \sum_k q_k \rho_k^h \mathbf{u}_k^h \quad (9)$$

where $q_e = Q_e/m_e$, and q_k is the mean charge per unit mass on the heavy particles of material k , which is simply related to Z_k^* . Clearly $q_k \rho_k^h$ is the total positive charge density of the heavy particles of composite material k , which can also be written as

$$q_k \rho_k^h = \sum_n q_k^n \rho_k^n \quad (10)$$

where $q_k^n = n|Q_e|/m_k^n$ is the charge per unit mass of X_k^n , and $\rho_k^n = \alpha_k \tilde{\rho}_k^n = \alpha_k m_k^n \tilde{n}_k^n$ is its partial density. Combining these relations with Eqs. (1) and (3), we obtain

$$q_k = \frac{\alpha_k}{\rho_k^h} |Q_e| \tilde{n}_k^e = \frac{\tilde{n}_k^e |Q_e|}{\tilde{\rho}_k^h} = \frac{Z_k^* |Q_e|}{m_k^h} \quad (11)$$

The velocities \mathbf{u}_k can now be obtained in terms of the \mathbf{u}_k^h by combining Eqs. (8), (9) and (11) to obtain

$$\rho_k \mathbf{u}_k = \rho_k^h \mathbf{u}_k^h + \frac{\rho_k^e}{\rho_e} \sum_j \left(\frac{m_j^0}{Z_j^* m_e} - 1 \right)^{-1} \rho_j^h \mathbf{u}_j^h \quad (12)$$

in which the second term clearly represents a small correction.

4. MOMENTUM EQUATIONS

In contrast to the continuity equations, it is necessary to write the momentum equations for the composite species *sans* electrons as already discussed. As noted in Ref. [3], these equations are readily obtained by assuming that all of the individual heavy species X_k^n ($n = 0, 1, 2, \dots$) contained within composite species k move with the same velocity \mathbf{u}_k^h . The individual momentum equations for those species are given by Eq. (11) of Ref. [3], and summing those equations over n for each k then yields

$$\begin{aligned} \frac{\partial(\rho_k^h \mathbf{u}_k^h)}{\partial t} + \nabla \cdot (\rho_k^h \mathbf{u}_k^h \mathbf{u}_k^h) &= -\nabla p_k^h + \rho_k^h (\mathbf{g} + q_k \mathbf{E}) + \sum_j \alpha_{kj} (\mathbf{u}_j - \mathbf{u}_k) \\ &+ \sum_j (\beta_{kj} - \beta_{jk}) \nabla \ln T + \beta_{ke} \nabla \ln T_e \end{aligned} \quad (13)$$

where \mathbf{E} is given by Eq. (9) of Ref. [3], and

$$\alpha_{kj} = \sum_{nm} \alpha_{kn,jm} \quad ; \quad \beta_{kj} = \sum_{nm} \beta_{kn,jm} \quad ; \quad \beta_{ke} = \sum_n \beta_{kn,e} \quad (14)$$

in which the coefficients $\alpha_{kn,jm}$, $\beta_{kn,jm}$, and $\beta_{kn,e}$ may in principle be evaluated using the relations given in Ref. [3]. Unfortunately, those relations involve the partial number densities $n_k^n = \alpha_k \tilde{n}_k^n$, which as previously discussed are not normally available. They could, if desired, be approximated by solving the relevant Saha equations [9,10], but this would entail a great deal of additional labor and complexity. This hardly seems justified, since the results would be approximate in any case and moreover would be unlikely to be consistent with the approximations used in constructing the equations of state, particularly since the Saha equations come in a variety of forms and remain controversial for two-temperature plasmas

[10–20]. We therefore adopt a simpler alternative approximation suggested by Chang [21], in which we disregard Eq. (14) and proceed as though composite species k *sans* electrons were actually a single species of identical particles with number density n_k^h , mass m_k^h , and charge $Q_k = m_k^h q_k = Z_k^* |Q_e|$. The coefficients α_{kj} , β_{kj} , and β_{ke} in Eq. (13) can then simply be evaluated using Eqs. (12)–(15), (17), (18) or (19), and (21) of Ref. [3].

Strictly speaking, Eq. (13) should also contain source or sink terms representing the electron momentum gained or lost by the heavy particles due to the bound electrons gained or lost during ionization or recombination. These terms are proportional to the corresponding source terms in the continuity equations for ρ_k^h , which are not however available since we do not solve those equations, as previously discussed. Fortunately, these source terms are very small due to the small value of m_e , so they will be neglected in Eq. (13) for simplicity.

Finally, we remark parenthetically that the *ad hoc* assumption that all the individual heavy species X_k^n contained within composite species k move with the same velocity is not strictly necessary, and could be relaxed without increasing the number of history variables that need to be advanced in time. This could be done by allowing those species to have different *velocities* \mathbf{u}_k^n while neglecting the differences between their *accelerations*. This is precisely the type of approximation upon which diffusional descriptions are based [1,2], and in the present context it would result in a diffusional description for the relative velocities $\mathbf{u}_k^n - \mathbf{u}_k^m$ *within* each composite species k while still computing the mean velocities \mathbf{u}_k^h dynamically. In essence, this would be a multifluid analog of the combined diffusion coefficient method of Murphy [22–24], and it would undoubtedly improve the accuracy of the description. However, although this approach would not increase the number of history variables, it would nevertheless greatly complicate the description, to the point where its

practicality might well be questioned. (Indeed, the reader may well feel that the description is already complicated enough, if not more so, as it stands.) Moreover, the more accurate approach would also require the use of state relations that provide all the number densities \tilde{n}_k^n , which as already discussed is not the case for most state routines or packages developed for real materials. In spite of its appeal, this approach therefore seems unsuitable for most practical applications at the present time, but it may warrant further consideration in the future.

5. SUMMARY OF EQUATIONS

The logical structure of the equations to be solved is as follows. The continuity equations (7) are used to determine the partial mass densities ρ_k of the composite species k by advancing these quantities forward in time. There is also at least one energy equation that determines the specific internal energy of the mixture as a whole [25], and possibly an electron energy equation that determines the internal energy and temperature of the free electrons. These densities and energies then serve as the independent thermodynamic variables in the thermodynamic state relations for the mixture, which are approximated in terms of the state relations for the “pure” composite species as discussed in Sect. 2 above. Whatever approximations are employed for this purpose then determine the volume fractions α_k and the temperature(s) T (and T_e), from which the $\tilde{\rho}_k$ and all other remaining thermodynamic variables, including the other “tilde” variables (number densities, pressures, etc.), may then be determined from the relations given in Sect. 2. At this point all quantities except the velocities \mathbf{u}_k^h have been advanced in time, and these advanced-time values are then available

for use in evaluating the various force terms in the momentum equations (13). Finally, these momentum equations are used to perform the time advancement of the velocities \mathbf{u}_k^h .

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