Plasma Chemistry and Plasma Processing, Vol. 11, No. 3, 1991

Ambipolar Diffusion in Multicomponent Plasmas

J. D. Ramshaw[1] and C. H. Chang[1]

Received June 4, 1990; revised January 25, 1991

A recently described self-consistent effective binary diffusion approximation is applied to ambipolar diffusion in a neutral multicomponent plasma in zero magnetic field, where forced diffusion due to the electric field **E** *plays an essential role. The field* **E** *is determined by the constraint that the net current flow produced by the diffusion fluxes must be zero. The resulting effective binary diffusion fluxes are the sum of those that would obtain for* $\mathbf{E} = 0$ *and ambipolar correction terms proportional to* **E**. *The formulation is self-consistent with respect to both mass and charge, the net diffusional fluxes of which are both identically zero. The results may be further simplified due to the small mass of the electrons. The effective binary diffusivity* D_e *of the electrons no longer appears in the simplified expressions. They are therefore well suited to numerical calculations, where the large value of* D_e *might otherwise have resulted in unacceptable stability or accuracy restrictions. The well-known effective doubling of ion diffusivities due to ambipolar diffusion occurs in simple situations but is not a general feature.*

KEY WORDS: Diffusion; ambipolar diffusion; multicomponent plasmas.

1. INTRODUCTION AND SUMMARY

In single-fluid descriptions of partially or fully ionized multicomponent plasmas, constitutive relations are in general required for the diffusional mass fluxes $\mathbf{J}_i$ of the individual components or species relative to the mass-averaged velocity of the plasma.[1,2] Forced diffusion due to the electric field **E** is an essential contribution to these fluxes. Usually **E** is not known *a priori* but is implicitly determined by the current density $\mathbf{J}_q$, which is known or determined on other grounds. Since $\mathbf{J}_q$ is a linear combination of the $\mathbf{J}_i$, **E** must assume the value necessary to make the $\mathbf{J}_i$ consistent with

[1] Idaho National Engineering Laboratory, EG&G Idaho, Inc., P.O. Box 1625, Idaho Falls, Idaho 83415

the given $\mathbf{J}_q$. Thus $\mathbf{E}$ plays the role of a parameter conjugate to $\mathbf{J}_q$, which serves as the mechanism by which constraints on $\mathbf{J}_q$ are enforced.

This paper is concerned with the case in which $\mathbf{J}_q = \mathbf{0}$, which is commonly referred to as ambipolar diffusion.[1,2] Ambipolar diffusion in simple situations is treated in textbooks. However, the resulting expressions are inadequate to deal with more complicated and realistic cases, such as detailed simulations of multicomponent thermal plasmas of arbitrary composition.[3,4] The growing interest in this and related problems creates a need for simple and tractable approximations for ambipolar diffusion in an arbitrary multicomponent plasma. Our purpose here is to derive such an approximation based on a recently described self-consistent effective binary diffusion approximation.[5] The resulting formulation is fully consistent with respect to both mass and charge, the net diffusional fluxes of which are both identically zero.

The present development is restricted to cases in which the magnetic field $\mathbf{B}$ is zero. This restriction is not essential or intrinsic to ambipolar diffusion, as $\mathbf{J}_q$ may vanish even when $\mathbf{B}$ does not. However, a proper treatment of diffusion in a magnetic field is considerably more complicated due to the velocity dependence of the magnetic forces.[6,7] This case falls within the purview of multicomponent magnetohydrodynamics, and is beyond the scope of this paper.

The development proceeds along the following outline. The general expressions of Ref. 5 are specialized to the present situation by identifying the specific body forces with gravity (which cancels out) and the electric forces on the charged species. The resulting effective binary diffusion fluxes $\mathbf{J}_i$ are the sum of those that would obtain if $\mathbf{E}$ were zero and additional terms proportional to $\mathbf{E}$. The corresponding current density $\mathbf{J}_q$ is then written as the appropriate linear combination of the $\mathbf{J}_i$, and $\mathbf{E}$ is determined by setting $\mathbf{J}_q = \mathbf{0}$. The resulting expression for $\mathbf{E}$ combines with the $\mathbf{J}_i$ to yield self-consistent effective binary ambipolar diffusion fluxes which automatically conserve both mass and charge.

The general expressions for $\mathbf{E}$ and the $\mathbf{J}_i$ may be further simplified due to the fact that the free electrons in the plasma are much lighter than the other components. The electrons dominate $\mathbf{E}$, but they otherwise contribute negligibly to the diffusion fluxes of the other species. The electron diffusion flux does not simplify in the same way, but it may easily be obtained from the condition that $\mathbf{J}_q = \mathbf{0}$. The net result is that the effective binary diffusivity of the electrons, D_e, no longer appears in the equations. This is fortunate, since D_e is very large and might otherwise give rise to severe stability and/or accuracy restrictions in numerical calculations. The resulting simplified formulation no longer exactly satisfies the constraint that the $\mathbf{J}_i$ sum to zero, but the discrepancy is negligible due to the small mass of the electrons. We also find that the well-known effective doubling of ion diffusivities due to ambipolar diffusion[1,2] is not a general feature, but occurs only in simple situations.

2. AMBIPOLAR DIFFUSION IN ARBITRARY MULTICOMPONENT PLASMAS

We begin with the self-consistent effective binary diffusion approximation of Ref. 5, which provides an explicit expression for the diffusional mass fluxes $\mathbf{J}_i$ relative to the mass-averaged velocity of the mixture. This expression may be written in the form

$$\mathbf{J}_i = -cM_iD_i\mathbf{G}_i + (\rho_i c/\rho)\sum_j M_jD_j\mathbf{G}_j \tag{1}$$

where $\mathbf{J}_i$, M_i, and ρ_i are, respectively, the diffusional mass flux, molecular weight, and partial mass density of species i, ρ is the total mass density (i.e., the sum of the ρ_i), c is the total molar concentration (i.e., the sum of ρ_i/M_i), D_i is the effective binary diffusivity of species i,[5] and the diffusional driving forces $\mathbf{G}_i$ are given by

$$\mathbf{G}_i = \mathbf{H}_i - (1/p)\left[\rho_i\mathbf{F}_i - (\rho_i/\rho)\sum_j \rho_j\mathbf{F}_j\right] \tag{2}$$

Here p is the pressure, $\mathbf{F}_i$ is the body force per unit mass acting on species i, and $\mathbf{H}_i$ represents the driving forces for concentration (ordinary), pressure, and thermal diffusion, namely

$$\mathbf{H}_i = \nabla x_i + (x_i - \rho_i/\rho)\nabla \ln p + K_i\nabla \ln T \tag{3}$$

where $x_i = \rho_i/(M_ic)$ is the mole fraction of species i and T is the temperature. The coefficients K_i are simply related to the species thermal diffusion coefficients, and they have the property that their sum over all species vanishes. It follows that both $\mathbf{G}_i$ and $\mathbf{H}_i$ also sum to zero in the same way.

In the present context, the only body forces present are gravity and the electric forces on the charged species. Thus $\mathbf{F}_i = \mathbf{g} + q_i\mathbf{E}$, where q_i is the charge per unit mass of species i, $\mathbf{E}$ is the electric field, and $\mathbf{g}$ is the acceleration of gravity. Equation (2) then becomes

$$\mathbf{G}_i = \mathbf{H}_i - (q_i\rho_i/p)\mathbf{E} \tag{4}$$

where use has been made of the neutrality condition

$$\sum_i q_i\rho_i = 0 \tag{5}$$

Notice that $\mathbf{g}$ has cancelled out and no longer appears. Combining Eqs. (1) and (4), we obtain

$$\mathbf{J}_i = -cM_iD_i\mathbf{H}_i + (\rho_i c/\rho)\sum_j M_jD_j\mathbf{H}_j + (c/p)\left[M_iq_i\rho_iD_i - (\rho_i/\rho)\sum_j M_jq_j\rho_jD_j\right]\mathbf{E} \quad (6)$$

in which the first two terms represent the species diffusion fluxes that would result in the absence of forced diffusion. This equation expresses the diffusion fluxes in terms of the $\mathbf{H}_i$, which are presumed known, and $\mathbf{E}$, which remains to be determined.

The current density in the plasma is related to the species diffusion fluxes by

$$\mathbf{J}_q = \sum_i q_i\mathbf{J}_i \quad (7)$$

Equation (7) actually defines the current density relative to the mass-averaged velocity of the plasma, but because of the neutrality condition there is no distinction between this current density and that in the laboratory frame.

In order to describe ambipolar diffusion, we must determine the electric field for which the current density vanishes. Combining Eqs. (5)-(7), setting $\mathbf{J}_q = \mathbf{0}$, and solving for $\mathbf{E}$, we obtain

$$\mathbf{E} = \frac{p\sum_i M_iq_iD_i\mathbf{H}_i}{\sum_i M_iq_i^2\rho_iD_i} \quad (8)$$

which may be referred to as the ambipolar electric field. This expression for $\mathbf{E}$, together with Eq. (6), defines our self-consistent effective binary diffusion approximation for ambipolar diffusion in multicomponent plasmas. The effective binary diffusion flux $\mathbf{J}_i$ is seen to be the sum of the flux that would result in the absence of forced diffusion and additional terms proportional to the ambipolar electric field $\mathbf{E}$. By construction, this approximation satisfies the condition $\mathbf{J}_q = \mathbf{0}$ as well as the constraint that the $\mathbf{J}_i$ properly sum to zero.[5]

3. SIMPLIFICATIONS DUE TO SMALL ELECTRON MASS

The free electrons in the plasma will be symbolically denoted by the species index $i = e$. They are much lighter than the other species, and their effective binary diffusivity D_e is consequently much larger. We are therefore led to examine the behavior of the general expressions for very small electron mass, i.e., very small M_e. For this purpose we may assume that D_e is proportional to $M_e^{-1/2}$.[1,2] It will therefore be convenient to introduce the small parameter $\varepsilon = M_e^{1/2}$. The orders of magnitude of the various quantities involved are then as follows: $q_e \sim \varepsilon^{-2}$, $\rho_e \sim \varepsilon^2$, $q_e\rho_e \sim 1$, $M_e q_e \sim 1$, $D_e \sim \varepsilon^{-1}$, and $M_e D_e \sim \varepsilon$, so that

$$M_e q_e^2 \rho_e D_e \sim M_e q_e D_e \sim \varepsilon^{-1} \tag{9}$$

and

$$M_e q_e \rho_e D_e \sim \varepsilon \tag{10}$$

First we examine the behavior of **E**. By virtue of Eq. (9), the electron terms dominate the sums in Eq. (8). Neglecting terms of order ε and higher, we readily find

$$\mathbf{E} = (p/q_e\rho_e)\mathbf{H}_e \tag{11}$$

In the absence of pressure and temperature gradients $\mathbf{H}_e = \nabla x_e$, and Eq. (11) then reduces to an expression previously derived under less general circumstances.[2]

Next we examine the species fluxes $\mathbf{J}_i$ for $i \neq e$. By virtue of Eq. (10), the electron terms in Eq. (6) are now of order ε *smaller* than the other terms. Again neglecting terms of order ε and higher, we obtain

$$\begin{aligned}\mathbf{J}_i = -cM_iD_i\mathbf{H}_i + (\rho_i c/p) \sum_{j \neq e} M_jD_j\mathbf{H}_j \\ + (c/p)\left[M_iq_i\rho_iD_i - (\rho_i/p) \sum_{j \neq e} M_jq_j\rho_jD_j\right]\mathbf{E} \qquad (i \neq e)\end{aligned} \tag{12}$$

with **E** given by Eq. (11). The restrictions on the sums in Eq. (12) may be formally accomplished by setting $D_e = 0$ in Eq. (6) for $i \neq e$.

The diffusional flux $\mathbf{J}_e$ of the electrons requires special treatment because it is inherently of order ε^2 and vanishes to order unity. Setting $\mathbf{J}_e = \mathbf{0}$ would therefore be correct to order unity with regard to mass flux, but this would improperly neglect the essential contribution of the electrons to the current density. The latter contribution is $q_e\mathbf{J}_e$, which is of order unity and must be retained in order to satisfy the condition $\mathbf{J}_q = \mathbf{0}$ to order unity. Since q_e is of order ε^{-2}, this requires an expression for $\mathbf{J}_e$ correct to order ε^2. Such an expression could be derived directly from Eqs. (6) and (8), but a simpler equivalent procedure is to solve for $\mathbf{J}_e$ from the condition $\mathbf{J}_q = \mathbf{0}$; i.e.,

$$\mathbf{J}_e = -(1/q_e) \sum_{i \neq e} q_i\mathbf{J}_i \tag{13}$$

Equation (13) is of course exact if the correct $\mathbf{J}_i$ of Eq. (6) are used. Here, however, we obtain the $\mathbf{J}_i$ for $i \neq e$ from Eq. (12), which is only correct to order unity. Since q_e is of order ε^{-2}, Eq. (13) then gives $\mathbf{J}_e$ correct to order ε^2.

Equations (11)-(13) are the proper simplified expressions that result when the small mass of the electrons is taken into account. Since they do not represent all the $\mathbf{J}_i$ to the same order in ε, the total diffusional mass flux (i.e., the sum of the $\mathbf{J}_i$) is no longer identically zero as it should be,[5] but is rather of order ε^2. Thus the simplified formulation is not fully self-consistent with respect to mass conservation, which is the price paid for the simplification. However, since the electron mass is very small, the discrepancy should be negligible for most purposes.

Notice that D_e no longer appears in the simplified formulation. This is fortunate, since D_e is very large and might otherwise have resulted in unacceptable stability or accuracy restrictions in numerical calculations. The simplified formulation is therefore well suited for use in numerical simulations.[3,4]

It is often stated that when the ion and electron temperatures are equal (as assumed in the present development), the essential effect of ambipolar diffusion is to effectively double the ion diffusivities.[1,2,8,9] It is not apparent from the preceding results how or whether this occurs. The reason is that this doubling is not in fact a general feature of ambipolar diffusion, but rather occurs only in simple special cases. (Indeed, it is not even possible in general to define effective "ambipolar diffusion coefficients" in such a way that the diffusion fluxes formally reduce to their $\mathbf{E} = \mathbf{0}$ forms.) One such case is that of a three-component plasma produced by partial ionization of a single atomic species; i.e., $A \rightleftarrows A^+ + e^-$. This situation is commonly treated in textbooks. Of course, one would not ordinarily use the effective binary diffusion approximation in this context, where exact analytical results can be obtained without it. However, our purpose here is not to give an exact treatment but rather to examine the behavior of our approximate expressions in this special case.

The three-component plasma contains neutral atoms ($i = a$), positive ions ($i = p$), and electrons ($i = e$). The neutrality condition of Eq. (5) reduces to $q_p\rho_p = -q_e\rho_e$. Moreover, the ions and electrons carry equal but opposite charges, so that $M_p q_p = -M_e q_e$. These relations combine to imply that $x_p = x_e$, from which it follows that $x_a = 1 - 2x_p$. In the absence of pressure and temperature gradients $\mathbf{H}_i$ reduces to ∇x_i, so that $\mathbf{H}_p = \mathbf{H}_e = \nabla x_p$ and $\mathbf{H}_a = -2\nabla x_p$. These relations then combine with Eqs. (11) and (12) for $i = p$ to yield

$$\mathbf{J}_p = -2(c/\rho)[\rho_p M_a D_a + (\rho - \rho_p) M_p D_p]\nabla x_p \tag{14}$$

in which the factor of 2 represents the ambipolar doubling. The quantity in square brackets is proportional to a weighted average of D_a and D_p, and may be related to the atom-ion binary diffusivity D_{ap} via the definition of D_i, namely[5]

$$D_i = \frac{1 - w_i}{\sum_{j \neq i} x_j / D_{ij}} \tag{15}$$

Here D_{ij} is the binary diffusivity for the pair (i, j), and the w_i are normalized weighting factors employed in the effective binary diffusion approximation.[5] Combining Eqs. (14) and (15), neglecting terms involving $1/D_{ea}$ and $1/D_{ep}$ (which are very small because D_{ea} and D_{ep} are very large), and neglecting the electronic contribution to the mass density, we obtain

$$\mathbf{J}_p = -2(c^2/\rho)(1 + w_e) M_a M_p D_{ap} \nabla x_p \tag{16}$$

The conventional choice[5] $w_e = x_e$ would be inappropriate here because of the very small mass of the electrons. The latter should be reflected in a w_e value much less than unity, which can then be neglected in Eq. (16). The resulting $\mathbf{J}_p$ has precisely the form appropriate to a binary mixture of ions and neutrals alone with no electric forces,[10] but with D_{ap} replaced by $2D_{ap}$. The doubling is not an artifact of the effective binary diffusion approximation, as the same result may readily be obtained from the full multicomponent diffusion equations[10] under the same assumptions. It is clear from the preceding development that the origin of the doubling lies in the special relations peculiar to the present case, and is not in any sense a general feature. In order to describe ambipolar diffusion in more general situations, it is simply necessary to use the more general equations (6) and (8), or (11)-(13).

4. CONCLUDING REMARKS

We have presented a self-consistent effective binary diffusion approximation to ambipolar diffusion in multicomponent plasmas of arbitrary composition. The approximation explicitly expresses the diffusional mass fluxes in terms of the diffusional driving forces. It is fully self-consistent with respect to both mass and charge, the net diffusional fluxes of which are both identically zero. We have also presented a simplified formulation based on the fact that the electrons are much lighter than the other components.

The present development has assumed that all species, including the electrons, have the same common temperature T. A corresponding theory for unequal electron and ion temperatures would require more general starting expressions for the $\mathbf{J}_i$, but would proceed along basically the same outline.

We are currently using the present formulation, with the simplifications due to small electron mass, in our ongoing simulations of thermal plasma processes.[3,4] It is hoped that these results will also find useful applications in fusion plasmas[11] and astrophysical problems.[12-14]

ACKNOWLEDGMENTS

We are grateful to S. Peter Gary for a helpful conversation, and to L. D. Cloutman for calling our attention to Refs. 12–14. This work was performed under the auspices of the U.S. Department of Energy, contract number DE-AC07-76-IDO1570, supported by the Division of Engineering and Geosciences, Office of Basic Energy Sciences, DOE-OER.

REFERENCES

1. N. A. Krall and A. W. Trivelpiece, *Principles of Plasma Physics* (McGraw-Hill, New York, 1973).
2. M. Mitchner and C. H. Kruger, Jr., *Partially Ionized Gases* (Wiley-Interscience, New York, 1973).
3. J. D. Ramshaw and C. H. Chang, "Numerical Modeling of Thermal Plasma Processes," manuscript in preparation.
4. J. D. Ramshaw and C. H. Chang, "LAVA: A Computer Code for Two- or Three-Dimensional Multicomponent Fluid Flow with Chemical Reactions," Idaho National Engineering Laboratory report, in preparation.
5. J. D. Ramshaw, *J. Non-Equilib. Thermodyn.* **15**, 295 (1990).
6. S. Chapman and T. G. Cowling, *The Mathematical Theory of Non-Uniform Gases* (Cambridge University Press, Cambridge, 1960), Chapter 18.
7. R. S. Devoto, *Z. Naturforsch.* **24a**, 967 (1969).
8. V. E. Golant, A. P. Zhilinsky, and I. E. Sakharov, *Fundamentals of Plasma Physics* (Wiley, New York, 1980).
9. H. V. Boenig, *Plasma Science and Technology* (Cornell University Press, Ithaca, New York, 1982).
10. R. B. Bird, W. E. Stewart, and E. N. Lightfoot, *Transport Phenomena* (Wiley, New York, 1960).
11. J. P. Freidberg, *Rev. Mod Phys.* **54**, 801 (1982).
12. L. H. Aller and S. Chapman, *Astrophys. J.* **132**, 461 (1960).
13. P. D. Noerdlinger, *Astron. Astrophys.* **57**, 407 (1977).
14. J. Wambsganss, *Astron. Astrophys.* **205**, 125 (1988).