THERMODYNAMIC ANALYSIS OF DIFFUSION COEFFICIENTS

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An analysis is presented of the process of diffusion of a multicomponent fluid through a solid porous medium. It is shown that there are conditions in which the diffusion fluxes vanish, and for which the multicomponent system may be replaced by a one-component system in aerodynamic problems.

Analysis of the flow of a single-phase multicomponent fluid with the aid of the thermodynamics of irreversible processes shows that diffusion, which is an irreversible process, is observed in the stream. Diffusion causes the production of entropy in the system, which may be a fluid only, or a mixture of a solid and a fluid, as in the case of flow of a fluid through solid porous media. The first kind of system has been examined in detail by De Groot [1], and the second—by the author, in [2–4]. The author also showed that this kind of analysis may easily be modified for a system consisting of only one fluid. Therefore, in the present article we will examine the flow of a multicomponent, single-phase fluid through a porous medium, and show that the concentration gradients in the fluid phase have an influence on the nature of the diffusion coefficients. With this objective we will make use of material from a previous article of the author's [4]. The reader should refer to it in order to become familiar with the explanation of the thermodynamic system and the derivation of the equations of continuity, momentum, energy, entropy, and entropy increase. We will employ the same notation in this article.

Thermodynamic system and entropy production. It is assumed that the thermodynamic system consists of a solid-fluid mixture, and that both phases are found at all points throughout the entire porous medium. The fluid phase may consist of n components. Its volume density, ρ, and the velocity of the center of mass, $\overline{V}$, are determined as follows:

$$\rho = \sum_{k=1}^{k=n} \rho_k, \quad \overline{V} = \frac{1}{\rho} \sum_{k=1}^{k=n} \rho_k \overline{V}_k, \tag{1}$$

where ρ_k and $\overline{V}_k$ are the volume density and the velocity, respectively, of component k. The diffusion m_k of component k is determined from the expression

$$\overline{m}_k = \rho_k (\overline{V}_k - \overline{V}). \tag{2}$$

From (1) and (2) we obtain

$$\sum_{k=1}^{k=n} \overline{m}_k = 0. \tag{3}$$

The mass concentration of component k is determined as

$$C_k = \rho_k / \rho. \tag{4}$$

For any extensive property of the fluid phase (or of any of its components), such as entropy, we may write

$$\frac{\partial \rho s}{\partial t} = \rho \frac{ds}{dt} - \operatorname{div} (\rho s \overline{V}), \tag{5}$$

where s is the specific entropy of the fluid phase. The total derivatives with respect to time of the properties of the parameters of the fluid and solid phases are calculated along their respective velocities, and therefore, for the fluid phase, or any of its components,

$$\frac{d}{dt} = \frac{\partial}{\partial t} + \overline{V} \operatorname{grad}. \tag{6}$$

Since the solid phase is stationary, the total and local derivatives with respect to time will be identical in this case. Assuming that there are no chemical reactions, we may obtain from the foregoing equations

$$\rho \frac{dC_k}{dt} = \rho \frac{\partial C_k}{\partial t} + \rho \overline{V} \operatorname{grad} C_k = -\operatorname{div} \overline{m}_k. \tag{7}$$

The author of [4] has shown that in unit volume of the system, the rate of production of entropy is

$$\sigma = \frac{1}{T} \left\{ \sum_i \sum_j (p\,\delta_{ij} - F_{ij}) \frac{\partial V_j}{\partial x_i} \right\} - \\ - \overline{q}\, \frac{\operatorname{grad} T}{T^2} - \sum_{k=1}^{k=n} \overline{m}_k \operatorname{grad} \frac{\mu_k}{T} - \frac{\overline{V}}{T} \overline{F}_f, \tag{8}$$

where T is the temperature, p is the volumetric pressure, δ_{ij} is the Kronecker delta, F_{ij} is a component of the volume stress, $\overline{q}$ is the total heat flux per unit area of the system, μ_k is the chemical potential of component k, and $\overline{F}_f$ is the friction force per unit volume of the system. Besides i and j, we may use x, y, z, and $x_x \equiv x$, $x_y \equiv y$ and $x_z \equiv z$.

Irreversible changes take place in the system due to viscous flow, heat conduction, diffusion, and motion of the center of mass of the fluid phase in the presence of a friction force. The first term is the scalar product of two second-order tensors, while the last three terms are scalar products of first-order tensors. As a result there is no connection between the first and the other three terms. But there may be, and it is assumed that there is, a connection between the last three terms. If σ'' is the entropy production per unit volume of the system due to these three terms, we obtain, from (3) and (8),

$$\sigma'' = -\bar{q}\,\frac{\operatorname{grad} T}{T^2} - \sum_{i=1}^{i=n-1} \bar{m}_i \operatorname{grad}\left(\frac{\mu_i - \mu_n}{T}\right) - \frac{\bar{V}}{T}\bar{F}_f, \quad (9)$$

where m_i and μ_i are, respectively, the diffusion and chemical potential of component i.

Phenomenological equations. From (9) we may write

$$\bar{q} = -L_{uu}\frac{\operatorname{grad} T}{T^2} - \sum_{i=1}^{i=n-1} L_{ui} \operatorname{grad}\left(\frac{\mu_i - \mu_n}{T}\right) - L_{uw}\bar{F}_f, \quad (10)$$

$$\bar{m}_p = -L_{pu}\frac{\operatorname{grad} T}{T^2} - \sum_{i=1}^{i=n-1} L_{pi} \operatorname{grad}\left(\frac{\mu_i - \mu_n}{T}\right) - L_{pw}\bar{F}_f, \quad (11)$$

$$\frac{\bar{V}}{T} = -L_{wu}\frac{\operatorname{grad} T}{T^2} - \sum_{i=1}^{i=n-1} L_{wi} \operatorname{grad}\left(\frac{\mu_i - \mu_n}{T}\right) - L_{ww}\bar{F}_f, \quad (12)$$

where $\overline{m}_p$ is the diffusion of component p ($p = 1, 2, 3, \ldots, n-1$).

The equations given above are called phenomenological equations, since they require experimental verification. In these equations the heat flux, the diffusion, and the motion of the center of mass of the fluid phase are regarded as "fluxes." It is assumed that each of these fluxes is linearly proportional to the gradient of temperature, the gradient of chemical potential, and to the friction force, which may be called the "forces." The flux and the force in each scalar product of (9) are conjugate quantities; for example, the heat flux is the conjugate of the temperature gradient. The connecting coefficients are known as the Onsager coefficients, being considered as constant, and not functions of the forces. If this is valid, we have

$$L_{wu} = L_{uw},\ L_{pu} = L_{up},\ L_{pi} = L_{ip},\ \text{etc.} \quad (13)$$

Expressions (13) are known as the Onsager reciprocal relations, and indicate how the various phenomena are related. These relations will be used widely in this analysis. According to the second law of thermodynamics

$$\sigma \geqslant 0 \text{ and } \sigma'' \geqslant 0. \quad (14)$$

It may therefore be shown that

$$L_{uu} \geqslant 0,\quad L_{ww} \geqslant 0,\quad L_{pp} \geqslant 0,\ \text{etc.}$$
$$L_{uu}L_{ww} - (L_{uw})^2 \geqslant 0,$$
$$L_{uu}L_{pp} - (L_{up})^2 \geqslant 0,\ \text{etc.} \quad (15)$$

Using the Gibbs-Duhem equation, we may also show that

$$\operatorname{grad}\left(\frac{\mu_i - \mu_n}{T}\right) = \left(\frac{h_n - h_i}{T^2}\right)\operatorname{grad} T + \left(\frac{v_i' - v_n'}{T}\right)\operatorname{grad} p' + \frac{1}{T}\sum_{k=1}^{k=n-1}\frac{\partial(\mu_i - \mu_n)}{\partial C_k}\operatorname{grad} C_k, \quad (16)$$

where h_i and V_i' are, respectively, the particular specific values of the enthalpy and the internal volume of component i, while p' is the internal pressure of the fluid phase.

The author has shown [3, 4] that, for moderate values of Reynolds number and negligibly small gravitation effects,

$$\bar{F}_f = r \operatorname{grad} p' = \operatorname{grad} p, \quad (17)$$

where r is the porosity of the system, and p is the volumetric pressure of the fluid phase.

Stationary states. The concept of stationary states was explained in detail by de Groot [1], and the author used it in his previous papers [3, 4]. It is based on the hypothesis that entropy production in the system always attains the least value consistent with the limits imposed upon it. By examining Eqs. (9)–(12), we see that σ'' may be written as a function of (n + 1) independent forces or variables. Any of these forces or variables may be "fixed" at a constant value, or left "free." If all the forces are free, there is no restraint on the system, and the result is a zero value of σ'', because of evolution of the system. Such a state is known as a stationary state of zero order. If we fix one variable and leave the remaining n free, the quantity σ'' attains its least value, commensurate with this restraint, and we obtain as a result a stationary state of the first order. Thus, the order is determined by the number of fixed independent variables. For a given order of stationary state, the condition of least value of the quantity σ'', together with the use of the Onsager reciprocal relations, leads to disappearance of the fluxes that are conjugate to the free forces (or variables). For example, in a stationary state of order one, in which the temperature gradient is fixed, the diffusion flows and the motion of the center of mass of the fluid phase vanish.

It should be noted here that once the validity of the entropy production equation and of the phenomenological equations is assumed, any stationary state described by these equations is permissible.

Characteristics of the diffusion coefficients. The Onsager coefficients in the diffusion Eq. (11) may be called diffusion coefficients. The nature of these coefficients will be examined from the viewpoint of the concept of stationary states. We will examine a stationary state of order two in which

$$\operatorname{grad} T/T^2 = \text{const}, \quad (18)$$

$$\bar{F}_f = \text{const} \quad (19)$$

and grad $[(\mu_i - \mu_n)/T]$ for all n − 1 components remains free. Evolution of this stationary state gives

$$\bar{m}_p = 0 \quad (p = 1, 2, 3, \ldots, n-1). \quad (20)$$

It may be seen from (17) and (19), that, by assigning constant $\bar{F}_f$, we obtain a constant pressure gradient. Since the velocity of the center of mass of the fluid phase is not zero, from (7) and (20) we obtain

$$\operatorname{grad} C_i = 0 \quad (i = 1, 2, 3, \ldots, n-1), \quad (21)$$

and so the fluid behaves like a one-component fluid. If we substitute (16), (17), (20) and (21) into (11), we obtain (n − 1) diffusion equations, determining the ratio of the pressure and temperature gradients, which is impossible, because of the initial conditions (18) and (19). Therefore, all the diffusion coefficients in the (n − 1) diffusion equations must be zero. Here it should be stressed that (21) is the key equation leading to this conclusion. If, for example, grad $C_i \neq 0$ (i = 1, 2, 3, ..., n − 1), then the values of all the diffusion coefficients could be determined from the (n − 1) diffusion equations. Therefore, when (21) is valid, we have

$$L_{iu}=L_{ui}=0,\quad L_{iw}=L_{wi}=0,\quad L_{ip}=L_{pi}=0, \quad (22)$$

where i = 1, 2, 3, ..., n − 1 and p = 1, 2, 3, ..., n − 1.

In order to determine in greater detail the dependence of the diffusion coefficients on the concentration gradients, we will examine a stationary state of n-th order, in which

$$\operatorname{grad} T/T^2=\text{const},$$

$$\bar{F}_f=\text{const},$$

$$\operatorname{grad}\left(\frac{\mu_i-\mu_n}{T}\right)=\text{const}\quad (i=2,3,4,\ldots,n-1) \quad (23)$$

and grad $(\mu_i - \mu_n)/T$ is left free. In this case there is no diffusion of the first component. From (11) we have

$$\bar{m}_1=-L_{1u}\frac{\operatorname{grad} T}{T^2}-$$

$$-\sum_{i=1}^{i=n-1} L_{1i}\operatorname{grad}\left(\frac{\mu_i-\mu_n}{T}\right)-L_{1w}\bar{F}_f=0, \quad (24)$$

and Eqs. (17) and (10) show that the value of the pressure gradient is fixed. Since $\bar{V}$ is not equal to zero, we obtain, from (7) and (24),

$$\operatorname{grad} C_1=0. \quad (25)$$

It may be seen from (16)–(19) that (23) gives us (n − 2) conditions for determining the concentration gradients for the (n − 2) components of the fluid phase. Thus, Eqs. (18), (19), (23) and (25) determine the values of all the forces in (24). Since these values may be arbitrary, Eq. (24) will be valid only when all the coefficients appearing in it are zero. We again note that (25) is the key equation leading to this conclusion. If grad C_1 were not zero, its value would be determined from (24) in terms of the diffusion coefficients. Thus, when grad C_1 equals zero,

$$L_{1u}=L_{u1}=0,\quad L_{1w}=L_{w1}=0,$$

$$L_{1i}=L_{i1}=0\ (i=1,2,3,\ldots,n-1). \quad (26)$$

In a similar way it may be shown that when grad C_2 equals zero

$$L_{2u}=L_{u2}=0,\quad L_{2w}=L_{w2}=0,$$

$$L_{2i}=L_{i2}=0\ (i=1,2,3,\ldots,n-1). \quad (27)$$

Generalizing Eqs. (22), (26) and (27), we obtain

$$L_{pu}=L_{up}=0,\quad L_{pw}=L_{wp}=0,\quad L_{pp}=0, \quad (28)$$

if grad C_p = 0 (p may have any of the following values: 1, 2, 3, ..., n − 1), and

$$L_{pi}=L_{ip}=0, \quad (29)$$

if grad C_p or grad C_i equals zero (i and p equal to any of the values 1, 2, 3, ..., n − 1 and i ≠ p).

We will now examine the case when the diffusion coefficients are continuous functions of the concentration gradients. To simplify the analysis, we assume that the fluid phase consists of two components. Then Eqs. (9)–(12) may be rewritten in the following form:

$$\sigma''=-\bar{q}\,\bar{x}_u-\bar{m}_1\bar{x}_m-\frac{\bar{V}}{T}\bar{F}_f, \quad (30)$$

$$\bar{q}=-L_{uu}\bar{x}_u-L_{u1}\bar{x}_m-L_{uw}\bar{F}_f, \quad (31)$$

$$\bar{m}_1=-L_{1u}\bar{x}_u-L_{11}\bar{x}_m-L_{1w}\bar{F}_f, \quad (32)$$

$$\bar{V}/T=-L_{wu}\bar{x}_u-L_{w1}\bar{x}_m-L_{ww}\bar{F}_f, \quad (33)$$

where

$$\bar{x}_u=\frac{\operatorname{grad} T}{T^2};\quad \bar{x}_m=\operatorname{grad}\left(\frac{\mu_1-\mu_2}{T}\right). \quad (34)$$

In this case a stationary state of second order, where the values of $\bar{x}_m$ and $\bar{F}_f$ are fixed, while the value of $\bar{x}_u$ remains free, leads to a zero value of $\bar{q}$.

We now suppose that the diffusion coefficients in (32) are continuous scalar functions of the concentration gradient and may be represented in the form

$$L_{1u}=L_{u1}=f_u(\bar{x}_1),\quad L_{1w}=L_{w1}=f_w(\bar{x}_1),$$

$$L_{11}=f_m(\bar{x}_1), \quad (35)$$

where

$$\bar{x}_1=\operatorname{grad} C_1. \quad (36)$$

From (30)–(33) and (35) we obtain

$$\sigma''=L_{uu}\bar{x}_u\bar{x}_u+2f_u(\bar{x}_1)\bar{x}_m\bar{x}_u+2L_{uw}\bar{F}_f\bar{x}_u+$$

$$+2f_w(\bar{x}_1)\bar{x}_m\bar{F}_f+f_m(\bar{x}_1)\bar{x}_m\bar{x}_m+L_{ww}\bar{F}_f\bar{F}_f. \quad (37)$$

Now, with fixed values of $\bar{x}_m$ and $\bar{F}_f$ and free $\bar{x}_u$, as a result of evolution of the system, we obtain

$$d\sigma''=2\{L_{uu}\bar{x}_u+f_u(\bar{x}_1)\bar{x}_m+L_{uw}\bar{F}_f\}\,d\bar{x}_u+$$

$$+\{2\bar{x}_u\,df_u(\bar{x}_1)+2\bar{F}_f\,df_w(\bar{x}_1)+\bar{x}_m\,df_m(\bar{x}_1)\}\bar{x}_m=0, \quad (38)$$

which, together with Eqs. (31) and (35) gives

$$2\bar{q}\,d\bar{x}_u+\{2\bar{x}_u\,df_u(\bar{x}_1)+2\bar{F}_f\,df_w(\bar{x}_1)+\bar{x}_m\,df_m(\bar{x}_1)\}\bar{x}_m=0. \quad (39)$$

From (16), (17), (34) and (36) we may write

$$\bar{x}_m=(h_2-h_1)\bar{x}_u+\left(\frac{v_1'-v_2'}{rT}\right)\bar{F}_f+\frac{1}{T}\frac{\partial(\mu_1-\mu_2)}{\partial C_1}\bar{x}_1, \quad (40)$$

and, since the values of $\bar{x}_m$ and $\bar{F}_f$ are fixed,

$$(h_2-h_1)\,d\bar{x}_u+\frac{1}{T}\frac{\partial(\mu_1-\mu_2)}{\partial C_1}\,d\bar{x}_1=0. \quad (41)$$

Therefore the differential of $f(\bar{x}_1)$ is not equal to zero, and the quantity taken into the parentheses (using which we obtain the scalar product $\bar{x}_m$ in (39)) is not zero. Thus, the quantity $\bar{q}$ in (39) is not zero. Since the quantity $\bar{q}$ for the stationary state examined must equal zero, Eq. (39) cannot be true. Thus we see that (35) is incorrect, and the diffusion coefficients cannot be continuous functions of the concentration gradients. Therefore, these coefficients are constants (possibly even zero) at the final values of the concentration gradients, and they can go to zero discretely at zero values of the concentration gradients. Together with Eqs. (15), (28) and (29), this conclusion may be written as follows:

$$L_{pp} \geqslant 0, \quad L_{pu} = L_{up} \gtreqless 0, \quad L_{pw} = L_{wp} \gtreqless 0, \tag{42}$$

if grad $C_p \neq 0$ ($p = 1, 2, 3, \ldots, n-1$):

$$L_{pp} = L_{pu} = L_{up} = L_{pw} = L_{wp} = 0, \tag{43}$$

if grad $C_p = 0$ ($p = 1, 2, 3, \ldots, n-1$);

$$L_{pi} = L_{ip} \gtreqless 0, \tag{44}$$

if grad $C_p \neq 0$ and grad $C_i \neq 0$ (i and $p = 1, 2, 3, \ldots, n-1$ and $i \neq p$)
and

$$L_{pi} = L_{ip} = 0, \tag{45}$$

if grad $C_p = 0$ or grad $C_i = 0$ (i and $p = 1, 2, 3, \ldots, n-1$ and $i \neq p$).

The equations given above describe the characteristics of the diffusion coefficients and their dependence on the concentration gradients.

If the system consists only of a fluid (i.e., the porosity r is equal to 1), then there is no friction force $\bar{F}_f$, and it therefore does not enter into the entropy production Eqs. (8) and (9). In other words, the motion of the center of mass of the fluid will be reversible. The result of this is that there will be no friction force terms in (10), (11), and (12). Obviously, the Onsager coefficients L_{ww}, L_{wu}, $L_{wi}, \ldots$ drop out, and there will be no differences between the volumetric and the internal properties. Equation (17) also will not be valid. Taking this into account, it may easily be shown that the nature of the diffusion coefficients in such a system also depends on the equations presented.

Sphere of application. We will examine a stationary state of the first order, in which $\bar{F}_f$ (or, in accordance with (17), the pressure gradient) is fixed at a constant value, while all the other forces remain free. As the author has shown [4], this stationary state may be used to calculate the Joule-Thomson coefficient. In this case the diffusion fluxes and the total heat flux are equal to zero, and Eq. (21) is applicable. Therefore, according to (22) or (43) and (45), all the diffusion coefficients are zero. Thus, the fluid behaves like a one-component fluid. From (10) and (17), we obtain

$$\bar{q} = -L_{uu}\frac{\operatorname{grad} T}{T^2} - rL_{uw}\operatorname{grad} p' = 0, \tag{46}$$

whence we may write

$$rT^2\frac{L_{uw}}{L_{uu}} = \left(\frac{\partial T}{\partial p'}\right)_{\bar{q},\, \bar{m}_1,\, \bar{m}_2,\, \ldots,\, \bar{m}_{n-1} = 0}. \tag{47}$$

Equation (47) in fact determines the Joule-Thomson coefficient. It should be noted that Eq. (53) in the author's previous article [4] requires alteration in view of the validity of the above equations.

If the system consists only of a fluid, the pressure gradient will not enter into (46). In such a system, where only the pressure gradient is constant, we have a stationary state of the first order, where the flow of the fluid is isothermal, and there is no diffusion, i.e., we have flow of a one-component fluid. It is evident that in this case no Joule-Thomson effect will be observed.

It should be noted that in aerodynamics we assume a condition leading to absence of diffusion and to a zero concentration gradient, while the air is regarded as a one-component fluid. Now we may easily show that in fact this condition is satisfied in all problems relating to the flow of air, when restraints are imposed on the pressure and the temperature gradients, or only on the pressure gradients, but not on the concentration gradients (or on the gradients of chemical potential). The evolution of the stationary states corresponding to these restraints, leads to zero diffusion flows and zero concentration gradients. If we first take the zero concentration gradients (as in the case of problems relating to flow of air) from (43) and (45), the diffusion fluxes at first will also be zero. In this case the above-mentioned stationary states were developed at the very beginning, and therefore the fluid behaves as a one-component fluid, independently of whether the flow is or is not steady.

We will now examine the possibility of a given stationary state being absent. For example, when the pressure and temperature gradients have been fixed [Eqs. (18) and (19)], we have a stationary state in which the fluid flows like a one-component fluid [Eqs. (20) and (21)]. This picture of the flow is the case of the ordinary momentum transfer between the components of the fluid. If under such restraints it is observed that the motion $\bar{V}_i$ of any component i differs from the motion $\bar{V}$ of the center of mass, this corresponds to the case when momentum transfer is impossible between the i-th component and the rest of the fluid. In this case we do not get the above-mentioned stationary state, and therefore Eqs. (9)–(12) which describe it, will not pertain. In this case it will be necessary to construct another model of the thermodynamic system by taking into account the total derivatives with respect to time [Eqs. (5) and (6)] of the properties of component i along its velocity $\bar{V}_i$, and the total derivatives with respect to time of the properties of the rest of the fluid phase along its motion. Thus, the fluid part of the system (in the case of a mixture, the solid-fluid) will be considered as a mixture of two "fluids," i.e., the "fluid" i and the remaining "fluid." This alters the character of the entropy production equation and of the phenomenological equations. This kind of model was used by Prigogine and Mazur [5, 6] in analyzing the behavior of liquid helium. As these authors showed, the method works if there is a resistance to the transfer of momentum between the components of the fluid.

In an experimental apparatus the pressure and temperature gradients may easily be controlled. Therefore the so-called stationary state may easily be altered and used as a criterion of the validity of the given analysis.

In accordance with what has been discussed, the solid and fluid phases in the investigation mentioned were examined separately, and not as a single entity.

As a result, the total derivatives with respect to time [Eqs. (5) and (6)] of the properties of the liquid phase (or of any of its components) were examined along $\overline{V}$, while the derivatives of the solid phase were examined along its velocity, which was zero in our case.

SUMMARY

The character of the diffusion coefficients is determined completely by Eqs. (42)–(45). These equations are valid for a system which may be a mixture of a solid and a fluid, or a fluid only. With the aid of these equations the conditions may easily be demonstrated under which a multicomponent fluid behaves as a one-component fluid. In the limits of the analysis given, a stationary state with fixed pressure and temperature gradients should lead to a picture of the flow, when all the components of the fluid move with the same velocity and have zero concentration gradients. Therefore the existence of such a stationary state may be used as a criterion of the limits of the validity of the present investigation. The treatment presented is valid only for the problems examined within the framework of the present work.

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