11 Diffusion and External Driving Forces

11.1 Overview

Diffusing particles experience a drift motion in addition to random diffusion, when an external driving force is applied. Table 11.1 lists examples of driving forces. An electric field is the most common example of an external force and is treated in detail below. Another example is the gradient of the non-ideal part of the chemical potential, which we have considered already in the previous chapter on interdiffusion. The nature of and analytic expressions for the driving forces can be deduced from the thermodynamics of irreversible processes (see Chap. 12 and [1, 2]).

For many ionic solids (see Chaps. 26 and 27) the electrical conductivity results from the transport of ions rather than electrons as is the case in metals and semiconductors. When the charge carriers are ions in an electronically insulating crystal or glass, the ionic motion under the influence of an electric field is described by the *ionic conductivity*. The dc conductivity, σ_{dc}, relates the electrical current density, $\boldsymbol{j}_e$, via Ohm's law to the applied electric field $\boldsymbol{E}$:

$$\boldsymbol{j}_e = \sigma_{dc} \boldsymbol{E} \,. \tag{11.1}$$

If the ions in the material are labelled i $(= 1, 2, \ldots)$, the conductivity can be written in terms of mobilities u_i and charges q_i as

$$\sigma_{dc} = \sum_i C_i \mid q_i \mid u_i \,, \tag{11.2}$$

where C_i is the number density of ions of kind i. An appropriate comparison between mobility and diffusivity of ions is obtained via the Nernst-Einstein relation discussed below.

The interplay of electron currents and atomic fluxes is necessary in the consideration of *electromigration* in metals [5, 6]. In metals ions are screened by the conduction electrons. Then, the effective charge of an ion can be very different from the charge of the ionic cores. An electronic current also exerts a force on the atomic species. The origin of this force is the scattering of electrons at the ion cores and the associated momentum transfer. The coupling between electronic and atomic current in metals at elevated temperatures is of considerable technical relevance, because it is the origin of electromigration.

Table 11.1. Examples of driving forces for drift motion of atoms

Force	Expression	Remarks
Gradient of electrical potential $\boldsymbol{E} = -\boldsymbol{\nabla} U$	$q^* \boldsymbol{E}$	q^*: effective electric charge
Gradient of chemical potential (non-ideal part)	$-\boldsymbol{\nabla}\mu$	μ: chemical potential
Temperature gradient $\boldsymbol{\nabla} T$	$-(Q^*/T)\boldsymbol{\nabla} T$ or $-S\nabla T$	Q^*: heat of transport S: Soret coefficient
Stress gradient	$-\boldsymbol{\nabla} U_{el}$	U_{el}: elastic interaction energy due to stress field
Gravitational force	mgz	m: particle mass g: acceleration due to gravity
Centrifugal force	$m^*\omega^2 r$	m^*: effective atomic mass ω: angular velocity r: distance from rotation axis

The latter is a major reason for the degradation of metallic interconnects in microelectronic devices.

Temperature gradients in a material can also act as driving force on diffusing atoms. The resulting effect is called *thermotransport* (also *thermomigration*). If there are simultaneous gradients of temperature and of concentration, we can combine Fick's first law and thermotransport to give

$$j = -D\frac{\partial C}{\partial x} - S\frac{\partial T}{\partial x}\,. \tag{11.3}$$

A steady state (index ss) can be established for $j = 0$. We then have

$$\left(\frac{\partial C}{\partial x}\right)_{ss} = -\frac{S}{D}\left(\frac{\partial T}{\partial x}\right)_{ss}\,. \tag{11.4}$$

This equation describes the concentration gradient established by thermotransport. This effect is called the *Ludwig-Soret effect* or often just the *Soret effect*. The Soret coefficient, S, and the heat of transport, $Q^* \equiv S/T$, may be of either sign, whereas D is always positive. Thermotransport is a relatively complex phenomenon since the system is, *per se*, not isothermal. For a more detailed discussion the reader is referred to [3–5].

In the case of *non-ideal alloys* the gradient of the chemical potential gives rise to a driving force. The gradient of the chemical potential can be expressed as a sum of two terms: the first term contains the concentration gradient and the second term the gradient of the activity coefficient. This case is discussed in detail in Chaps. 10 and 12.

A uniform stress field cannot generate a particle flux. However, a driving force can arise from the *stress gradient*. Its effects must be considered whenever the interaction energy of the particle with the stress field is large enough. Examples are the *Gorski effect*, which can be observed for hydrogen diffusion in metals (see Chap. 14), and the formation of *Cottrell atmospheres* in the stress field of dislocations [7].

Gravitational forces are weak and accordingly play no rôle in solid-state diffusion. In gases and liquids, however, gravitational forces can cause sedimentation effects. The experimental observation of effects of *centrifugal forces* requires rapid rotation. Equilibrium sedimentation of Au in solid sodium and lead has been studied in centrifuge experiments [8]. The results have been discussed to demonstrate the interstitial nature of fast diffusers.

11.2 Fick's Equations with Drift

Suppose that an *external driving force* F acts on diffusing particles. After a short transition period, a steady state particle flux develops. The *drift velocity* $\bar{v}$ of the particles under the action of the driving force is

$$\bar{v} = uF \,. \tag{11.5}$$

u is called the *mobility*. The mobility is the drift velocity for a unit driving force, i.e. for $F = 1$. The particle flux is $C\bar{v} = CuF$. The total flux due to diffusion plus the action of the driving force is

$$j = -D\frac{\partial C}{\partial x} + \bar{v}C \,. \tag{11.6}$$

The first term is the well-known Fickian term and the second term is the drift term. Thermodynamics of irreversible processes shows that the distinction between these two terms is more practical than fundamental (see Chap. 12). Formally, the second term in Eq. (11.6) is also a flux which is proportional to a gradient. In the case of an electrical driving force, this is the electrical potential gradient. If we combine Eq. (11.6) with the continuity equation (2.4), we get

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x}\left(D\frac{\partial C}{\partial x}\right) - \frac{\partial}{\partial x}(\bar{v}C). \tag{11.7}$$

If the driving force and, consequently, the drift velocity $\bar{v}$ and the diffusion coefficient are independent of x, Eq. (11.7) reduces to

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - \bar{v}\frac{\partial C}{\partial x} \,. \tag{11.8}$$

Then, it is possible to reduce the problem of solving the differential equation (11.8) by applying the following transformation [9, 10]

$$C = C^* \exp\left(\frac{\bar{v}}{2D}x - \frac{\bar{v}^2 t}{4D}\right) . \tag{11.9}$$

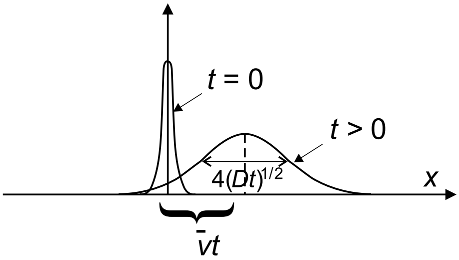

Fig. 11.1. Schematic illustration of diffusion and drift

Substitution of Eq. (11.9) in Eq. (11.8) yields a linear differential equation for C^*

$$\frac{\partial C^*}{\partial t} = D \Delta C^* \,. \tag{11.10}$$

Hence solutions for Eq. (11.8) are available, if Eq. (11.10) can be solved for the given boundary condition.

The concentration field $C(x,t)$ originating from a thin-film diffusion source is modified by a constant driving force. The appropriate diffusion equation is Eq. (11.8). Its solution can be obtained by inserting the thin-film solution without drift, Eq. (3.9), into Eq. (11.9). We get

$$C(x,t) = \frac{N}{2\sqrt{\pi D t}} \exp\left[-\frac{(x - \bar{v}t)^2}{4Dt}\right] . \tag{11.11}$$

This corresponds to a Gaussian distribution, the center of which shifts with the velocity $\bar{v}$. Equation (11.11) somehow justifies the distinction between the Fickian term and the drift term in Eq. (11.6). The first term leads to a broadening of the distribution of diffusing particles and the second term causes a translation of the whole distribution (Fig.11.1).

The fact that diffusion leads to a Gaussian distribution of the diffusing particles reveals their underlying random movement, whereas under the influence of an external force an additional directed motion occurs. On the other hand, both processes involve the same elementary jumps of the particles. This indicates that D and $\bar{v}$ must be related to each other. This relation is known as the *Nernst-Einstein relation.* Its derivation and some of its implications are considered in the following.

11.3 Nernst-Einstein Relation

We consider a system with one mobile component, where the flux resulting from an external driving force exactly counterbalances the diffusion flux. The combined effect of a concentration gradient and of a driving force can lead to a steady state, if the corresponding fluxes are equal and opposite in sign, i.e. if the total flux vanishes. Then, we get from Eq. (11.6)

$$0 = -\tilde{D}\frac{\partial C}{\partial x} + \bar{v}C\,. \tag{11.12}$$

The diffusion coefficient $\tilde{D}$ in Eq. (11.12) refers to a chemical composition gradient as will become evident below. It is definitely conceived as a *chemical diffusion coefficient* not as a tracer diffusion coefficient. Lack of appreciation of this fact leads to misunderstandings and inconsistencies.

Let the diffusing substance be contained in a cylinder and let us suppose that $\bar{v} = uF$ is the stationary velocity in negative x-direction due to an external field. Then, the solution of Eq. (11.12) is

$$C = C_0 \exp\left(-\frac{\bar{v}}{\tilde{D}}x\right)\,, \tag{11.13}$$

where C_0 denotes the stationary concentration at $x = 0$. Let us further assume that the external force is the derivative of a potential U:

$$F = -\frac{\partial U}{\partial x}\,. \tag{11.14}$$

At thermodynamic equilibrium, the distribution of non-interacting particles must also follow the *Boltzmann distribution*[1]

$$C(x) = \alpha \exp\left(-\frac{U}{k_{\mathrm{B}}T}\right)\,, \tag{11.16}$$

where k_{B} denotes the Boltzmann constant, T absolute temperature, and α a constant. Differentiation with respect to x yields

$$\frac{\partial C}{\partial x} = -\frac{C}{k_{\mathrm{B}}T}\frac{\partial U}{\partial x} = \frac{CF}{k_{\mathrm{B}}T}\,. \tag{11.17}$$

Substituting this equation in Eq. (11.12), we get

$$\tilde{D} = \frac{\bar{v}}{F}k_{\mathrm{B}}T = uk_{\mathrm{B}}T = u\frac{RT}{N_A}. \tag{11.18}$$

$R = k_{\mathrm{B}}N_A$ denotes the gas constant and N_A the Avogadro number. Equation (11.18) relates the chemical coefficient $\tilde{D}$ and the mobility u of the diffusing particles. This relation is called the *Nernst-Einstein* relation.

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[1] For a gas in the gravitational field of the earth the potential acting on gas molecules is $U = mgz$, where m is their mass, g the acceleration in the gravitational field, and z the height. Then we get

$$C = C_0 \exp\left(-\frac{mgz}{k_{\mathrm{B}}T}\right)\,. \tag{11.15}$$

This is the well-known formula for the decrease of gas concentration in the atmosphere with increasing height.

11.4 Nernst-Einstein Relation for Ionic Conductors and Haven Ratio

Let us now suppose that the external driving force is the result of an electric field E acting on ions with charge q. Then we have

$$F = qE \,. \tag{11.19}$$

Using Eq. (11.18) the flux of ions in the electric field can be written as

$$j = \bar{v}C = \frac{qC\tilde{D}}{k_\mathrm{B}T}E \,. \tag{11.20}$$

The associated electric current density is

$$j_e = qj \ = \frac{q^2C\tilde{D}}{k_\mathrm{B}T}E \,. \tag{11.21}$$

This equation is Ohm's law, $j_e = \sigma_{dc}E$, with the ionic d.c. conductivity

$$\sigma_{dc} = \frac{q^2C\tilde{D}}{k_\mathrm{B}T} \,. \tag{11.22}$$

caused by the mobile ions. In the solid-state diffusion literature this relation is often called in a rather misleading way the Nernst-Einstein relation. However, we remind the reader that we used in its derivation Eq. (11.16) – an equation which holds for non-interacting particles only.

More generally, interactions are present between the particles. For this case, Murch [11] showed that the general form of the Nernst-Einstein relation is

$$\sigma_{dc} = \frac{q^2C\tilde{D}}{k_\mathrm{B}T}\left(\frac{\partial \ln N}{\partial \mu}\right) , \tag{11.23}$$

where μ is the chemical potential of the particles and N their site fraction.

When the distribution of particles is completely ideal (no interaction, not even site blocking effects), the thermodynamic factor is unity and Eq. (11.23) reduces to Eq. (11.22). In this special case, the diffusion coefficient $\tilde{D}$ equals the tracer diffusion coefficient D^*, so that

$$\sigma_{dc} = \frac{q^2CD^*}{k_\mathrm{B}T} \,. \tag{11.24}$$

When the particles are ideally distributed but subject to site blocking effects, Eq. (11.23) can be written as [11]

$$\sigma_{dc} = \frac{q^2CD^*}{fk_\mathrm{B}T} , \tag{11.25}$$

where f is the tracer correlation factor (see Chap. 4). This equation is appropriate to very dilute solutions. Ionic crystals having a virtually perfect lattice of ions fall into this category.

Charge diffusion coefficient: In the solid-state diffusion literature Eq. (11.22) is often used to calculate another 'diffusion coefficient' called the *charge diffusion coefficient* (sometimes denoted also as the *conductivity diffusion coefficient* or the *electrical diffusion coefficient*) via:

$$D_\sigma = \frac{k_{\mathrm{B}} T \sigma_{dc}}{C q^2} = \frac{R T \sigma_{dc}}{N_A C q^2} \,. \tag{11.26}$$

R denotes the gas constant and N_A the Avogadro number. D_σ has indeed the dimensions of a diffusion coefficient. However, it does not correspond to any diffusion coefficient that can be measured by way of Fick's laws. The identification of D_σ with a diffusion coefficient is only adequate for non-interacting particles.

What can be done is to use Eq. (11.26) as a definition of D_σ, recognising that at the same time D_σ has no Fickian meaning. It is, however, misleading in these circumstances to call Eq. (11.26) the Nernst-Einstein equation. Equation (11.26) is then used purely as a means of changing the d.c. conductivity σ_{dc} to a quantity that has the dimensions of a diffusion coefficient.

Haven ratio: It is common practice in solid-state ionics to define the so-called *Haven ratio* H_R, which is simply the ratio of the tracer diffusion coefficient, D^*, and the charge diffusion coefficient, D_σ:

$$H_R \equiv \frac{D^*}{D_\sigma} \,. \tag{11.27}$$

In view of the remarks about D_σ, it is appropriate to ask whether the Haven ratio has a straightforward physical meaning. Apart from some simple models (mentioned below) a general theory of H_R is not yet available.

For a *hopping model of ionic conduction*, provided that the ionic jumps are mediated by low concentrations of vacancies like in NaCl (see Chap. 26), one gets [11, 13]

$$H_R = \frac{f}{f_{AA}} \,. \tag{11.28}$$

Here f is the tracer correlation factor of the atoms and f_{AA} the physical or collective correlation factor (see Chap. 12). If one can neglect collective correlations, which is justified for an almost full or empty lattice of charge carriers, the interpretation of the Haven ratio is straightforward. This is the case for diffusion mechanisms such as the vacancy mechanism. Then, the Haven ratio simply equals the tracer correlation factor:

$$H_R \approx f \,. \tag{11.29}$$

In such a case, from a measurement of the Haven ratio the correlation factor and then the mechanism of diffusion can be exposed (see Chap. 7).

For a collective mechanism, such as the *interstitialcy mechanism*, the displacement of the charge is different from the displacement of the tracer atom. For a colinear interstitialcy mechanism the charge moves two jump distances whereas the tracer moves only one (see Chap. 6). The Haven ratio then becomes

$$H_R = \frac{f d^2}{f_{AA} d_q^2}, \tag{11.30}$$

where d and d_q are the jump distances of the tracer and of the charge, respectively.

A classic example for identifying the mechanism using experimental Haven ratios is the work of FRIAUF [12] on the motion of Ag in AgBr. It was observed that the Haven ratio varies from 0.5 at low temperatures to 0.65 at high temperatures. This behaviour of the Haven ratio was attributed to the simultaneous action of colinear and non-colinear interstitialcy jumps (see Chap. 26).

Much less satisfactorily interpreted are Haven ratios for fast ion conductors with highly disordered sublattices and for ion-conducting glasses. Haven ratios in such materials have been reviewed by MURCH [13] and by JAIN AND KANERT [14]. Further comments on the subject of correlations effects and ionic diffusion can be found in the review by MURCH [11] and the literature cited therein, as well as in a paper on Monte Carlo simulations of the Haven ratio of alkali ions in oxide glasses [15]. Pressure-dependent diffusivities and Haven ratios in alkali borate glasses are considered by IMRE ET AL. [18].

11.5 Nernst-Planck Equation – Interdiffusion in Ionic Crystals

Let us now consider two ionic system AX and BX with two ionic species A and B, which share the same sublattice of a crystal. Let us assume that only the species A and B are mobile, the X sublattice being fixed. During interdiffusion the two mobile species compete for vacancies on the common sublattice of A and B. Let us first consider the case of an ideal solution and the absence of an electric driving force. Then fluxes j_A and j_B are connected to the concentration gradients via

$$j_A = -D_A^* \frac{\partial C_A}{\partial x} \quad \text{and} \quad j_B = -D_B^* \frac{\partial C_B}{\partial x} \tag{11.31}$$

with

$$\frac{\partial C_A}{\partial x} = -\frac{\partial C_B}{\partial x}. \tag{11.32}$$

Since D_A^* and D_B^* are not equal, a net flux of electric charge $j_A \neq j_B$ would develop. However, the condition of electrical neutrality requires that this net flux must be zero.

Any electrical field E gives rise to drift fluxes. More general, the flux equations should then be written as

$$j_A = -D_A^* \frac{\partial C_A}{\partial x} + \frac{q C_A D_A^*}{k_\mathrm{B} T} E \quad \text{and} \quad j_B = -D_B^* \frac{\partial C_B}{\partial x} + \frac{q C_B D_B^*}{k_\mathrm{B} T} E \,. \tag{11.33}$$

Here we have introduced the drift term from the Nernst-Einstein equation and q denotes the (identical) charges of ions A and B. The condition of electrical neutrality requires

$$j_A + j_B = 0 \,. \tag{11.34}$$

It follows that

$$E = \frac{k_\mathrm{B} T}{q} \frac{D_A^* - D_B^*}{C_A D_A^* + C_B D_B^*} \frac{\partial C_A}{\partial x} \,. \tag{11.35}$$

We now recall the intrinsic diffusivities of Eq. (10.20), which we repeat for convenience:

$$j_A = -D_A^I \frac{\partial C_A}{\partial x} \quad \text{and} \quad j_B = -D_B^I \frac{\partial C_A}{\partial x} \,. \tag{11.36}$$

Inserting Eqs. (11.33) and (11.35), we arrive at:

$$D_A^I = D_B^I = \frac{D_A^* D_B^*}{N_A D_A^* + N_B D_B^*} \,. \tag{11.37}$$

This equation is called the *Nernst-Planck equation* for ideal solutions. For interdiffusion between ionic crystals AX and BX, there is no Kirkendall effect because there is no separation of charge and therefore no net vacancy flow. Equation (11.37) can also be written as

$$\frac{1}{D_A^I} + \frac{1}{D_B^I} = \frac{N_A}{D_B^*} + \frac{N_B}{D_A^*} \,. \tag{11.38}$$

For a non-ideal solution Eq. (11.37) must be multiplied by the thermodynamic factor Φ:

$$D_A^I = D_B^I = \frac{D_A^* D_B^*}{N_A D_A^* + N_B D_B^*} \Phi \equiv \tilde{D}_{Nernst-Planck} \,. \tag{11.39}$$

We mention without prove that in terms of the transport coefficients discussed in Chap. 12 one gets:

$$\tilde{D}_{Nernst-Planck} = \frac{2f}{1-f} \frac{L_{AB}}{N_A N_B} \,. \tag{11.40}$$

This equation shows that interdiffusion in ionic crystals is completely due to the off-diagonal term L_{AB} of the Onsager matrix.

11.6 Nernst-Planck Equation *versus* Darken Equation

The Nernst-Planck equation (11.39) and the Darken equation (10.34) differ significantly if the ratio of the tracer diffusivities is far from unity:

$$\tilde{D}_{Nernst-Planck} \neq \tilde{D}_{Darken} \,. \tag{11.41}$$

We have seen above that the interdiffusion coefficient described by the Nernst-Planck equation corresponds to a consecutive connection of the two diffusivities; it is controlled by the slower diffusing component. By contrast, the Darken expression is analogous to the paralled connection of the two diffusivities, which is controlled by the faster diffusing component.

In addition, we note that the Nernst-Planck and the Darken equations represent two limiting cases of interdiffusion in a binary metallic alloy:

We recall that the Darken equation has been derived under the assumption that vacancies are practically at thermal equilibrium during the interdiffusion process and that the vacancy flux compensates the unequal fluxes of the atomic species A and B. Usually, this is justified for a macroscopic diffusion sample, when sufficient sinks and sources for vacancies (e.g., dislocations) keep the vacancy concentration close to equilibrium in spite of non-vanishing vacancy fluxes.

Non-equilibrium vacancy distributions can be generated by the difference of the intrinsic fluxes of atoms, if the density and/or efficiency of vacancy sources/sinks are insufficient. Nazarov and Gurov have performed an analysis of interdiffusion in binary alloys taking into account non-equilibrium vacancies [16]. They have shown that the evolution of an interdiffusion profile is then governed by

$$\tilde{D}_{Nazarov-Gurov} = \frac{D_A^* D_B^*}{N_A D_A^* + N_B D_B^*} \Phi \equiv \tilde{D}_{Nernst-Planck} \,. \tag{11.42}$$

The Nernst-Planck expression for interdiffusion in ionic crystals and the Nazarov-Gurov expression for interdiffusion in a binary alloy with non-equilibrium vacancies are identical.

Furthermore, the interdiffusion coefficients of Darken and of Nazarov-Gurov correspond to different space and time regimes. This has been pointed out by Gusak and coworkers (see, e.g., [17]):

- The Darken expression, Eq. (10.34), governs the interdiffusion process for long diffusion times

$$t \gg \frac{\tau_V}{C_V} \,. \tag{11.43}$$

Here τ_V is the mean life-time of vacancies between their creation at vacancy sources and their annilhilation at vacancy sinks. C_V denotes the vacancy concetration. This condition is fulfilled when the average distance between vacancy sources and sinks, $\sqrt{D_V \tau_V}$, is small as compared to the width of the interdiffusion zone, i.e. for

$$\tilde{D}_{Darken}\, t \gg D_V \tau_V \,, \tag{11.44}$$

D_V denotes the vacancy diffusivity.

- The Nazarov-Gurov equation governs the interdiffusion process for short diffusion times

$$t \ll \frac{\tau_V}{C_V} \,. \tag{11.45}$$

This corresponds to

$$\tilde{D}_{Nazarov-Gurov}\, t \ll D_V \tau_V \,, \tag{11.46}$$

Then, the width of the interdiffusion zone is much smaller than the average distance between vacancy sources and sinks.

Loosely speaking, non-equilibrium vacancies and the Nazarov-Gurov equation are important on ‘nanoscopic scales’, whereas the Darken equation is relevant for interdiffusion on ‘macroscopic scales’.

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