Appendix A

Theoretical Estimation of Diffusion Coefficients for Binary Gas Mixtures

The diffusion coefficient D_{12} for the isothermal diffusion of *species 1* through constant-pressure binary mixture of *species 1* and *2* is defined by the relation

$$J_1 = -D_{12}\nabla c_1, \tag{A.1}$$

where J_1 is the flux of *species 1* and c_1 is the concentration of the diffusing species.

Mutual-diffusion, defined by the coefficient D_{12}, can be viewed as diffusion of *species 1* at infinite dilution through *species 2*, or equivalently, diffusion of *species 2* at infinite dilution through *species 2*. **Self-diffusion**, defined by the coefficient D_{11}, is the diffusion of a substance through itself.

There are different theoretical models for computing the mutual (self) diffusion coefficient of gases. For non-polar molecules, Lennard-Jones potentials provide a basis for computing diffusion coefficients of binary gas mixtures [30]. The mutual diffusion coefficient, in units of cm^2/s is defined as

$$D_{12} = 0.001858\ T^{3/2}\sqrt{\frac{M_1 + M_2}{M_1 M_2}}\frac{f_D}{p\sigma_{12}^2\Omega_D}, \tag{A.2}$$

where T is temperature of the gas in units of Kelvin; M_1 and M_2 are molecular weights of *species 1* and *2*; p is the total pressure of the binary mixture in units of atmospheres; f_D is the second-order correction, usually between 1.00 and 1.03; σ_{12} is the Lennard-Jones force constant for the gas mixture, defined by $\sigma_{12} = 1/2\ (\sigma_1 + \sigma_2)$; Ω_D is the collision integral defined by

$$\Omega_D = \frac{1.06036}{(T^*)^{0.15610}} + \frac{0.19300}{\exp(0.47635\ T^*)} + \frac{1.03587}{\exp(1.52996\ T^*)} + \frac{1.76474}{\exp(3.89411\ T^*)}, \tag{A.3}$$

where $T^* \equiv kT/\epsilon_{\circ 12}$, k is the Boltzman gas constant, $\epsilon_{\circ 12} = (\epsilon_{\circ 1}\epsilon_{\circ 2})^{1/2}$ and $\epsilon_{\circ 12} = \sqrt{\epsilon_{\circ 1}\epsilon_{\circ 2}}$.

Values of $\sigma_{1(2)}$, Ω_D and $\epsilon_{\circ 1(2)}$ are tabulated for most naturally occurring gases [30].

The self-diffusion coefficient of a gas can be obtained from Eq. A.2, by observing that for a one-gas system: $M_1 = M_2 = M$, $\epsilon_{\circ 1} = \epsilon_{\circ 2}$ and $\sigma_1 = \sigma_2$. Thus,

$$D_{11} = 0.001858\ T^{3/2}\sqrt{\frac{2}{M}}\frac{f_D}{p\sigma_{11}^2\Omega_D}. \tag{A.4}$$

It is useful to define **observable diffusion**, D_{obs}, which is diffusion that one observes in an experiment. Observable diffusion os *species 1* in the binary mixture of *species 1* and *species 2* is

$$\begin{aligned} \frac{1}{D_{obs,1}} &= \frac{p_1/(p_1+p_2)}{D_{11}(p=1atm)/(p_1+p_2)} + \frac{p_2/(p_1+p_2)}{D_{12}(p=1atm)/(p_1+p_2)} \\ &= \frac{p_1}{D_{11}(p=1atm)} + \frac{p_2}{D_{12}(p=1atm)} \\ &= \frac{1}{D_{11}(p=p_1)} + \frac{1}{D_{12}(p=p_2)}. \end{aligned} \tag{A.5}$$

Equation A.5 has a simple physical explanation when applied to gases. The observable diffusion rate of gas *1* in the mixture of gases *1* and *2* is equal to the diffusion rate of one atom of gas *1* through the rest of atoms of gas *1*, plus the diffusion rate of one atom of gas *1* through the atoms of gas *2*. Equation A.5 enables the estimation of the diffusion coefficient for the binary mixture of ^{129}Xe-nitrogen and ^{3}He-nitrogen.

A.0.1 Observable Diffusion Constant for a Mixture of Xe-129 and Nitrogen

The relevant parameters [30] are:

$$\sigma_{Xe} = 4.047 \quad \epsilon_{\circ Xe}/k = 231.0 \quad M_{Xe} = 130.4$$
$$\sigma_{N2} = 3.798 \quad \epsilon_{\circ N2}/k = 71.4 \quad M_{N2} = 28$$

At $T = (303 \pm 10)$ K and $p = (p_{Xe} + p_{N2})$ atm,

$$\begin{array}{llll} \sigma_{Xe-N2} = 3.9225 & \frac{\epsilon_{\circ Xe-N2}}{k} = 128.42 & \frac{kT}{\epsilon_{\circ Xe-N2}} = 2.398 & \Omega_D = 1.0183 \\ \sigma_{Xe-Xe} = 4.047 & \frac{\epsilon_{\circ Xe-Xe}}{k} = 231 & \frac{kT}{\epsilon_{\circ Xe-Xe}} = 1.333 & \Omega_D = 1.2696. \end{array} \tag{A.6}$$

The above parameter values yield

$$D_{Xe-N2} = \frac{0.1303 \times 10^{-4}}{(p_{Xe} + p_{N2})} \text{ m}^2/\text{s} \tag{A.7}$$
$$D_{Xe-Xe} = \frac{0.0584 \times 10^{-4}}{(p_{Xe} + p_{N2})} \text{ m}^2/\text{s}. \tag{A.8}$$

The observable diffusion rate for a mixture of ^{129}Xe and Nitrogen gas is therefore

$$\frac{1}{D_{obs}} = \frac{p_{Xe}}{0.0584 \times 10^{-4} \text{ m}^2/\text{s}} + \frac{p_{N2}}{0.1303 \times 10^{-4} \text{ m}^2/\text{s}}. \tag{A.9}$$

The cell used in Xenon experiments had the following pressures: $p_{Xe} = (0.48 \pm 0.01)$ atm and $p_{N2} = (0.14 \pm 0.01)$ atm. The theoretical estimation of the observable diffusion constant is thus $D_{obs} = (1.08 \pm 0.08) \times 10^{-5}$ m^2/s.

A.0.2 Observable Diffusion Constant for a Mixture of He-3 and Nitrogen

The relevant parameters [30] are:

$$\sigma_{He} = 2.551 \quad \epsilon_{\circ He}/k = 10.22 \quad M_{He} = 4$$
$$\sigma_{N2} = 3.798 \quad \epsilon_{\circ N2}/k = 71.4 \quad M_{N2} = 28$$

At $T = (308 \pm 10)$ K and $p = (p_{He} + p_{N2})$ atm,

$$\begin{array}{llll} \sigma_{He-N2} = 3.1745 & \frac{\epsilon_{\circ He-N2}}{k} = 27.013 & \frac{kT}{\epsilon_{\circ He-N2}} = 11.587 & \Omega_D = 0.7260 \\ \sigma_{He-He} = 2.551 & \frac{\epsilon_{\circ He-He}}{k} = 10.22 & \frac{kT}{\epsilon_{\circ He-He}} = 30.626 & \Omega_D = 0.6231. \end{array} \tag{A.10}$$

The above parameter values yield

$$D_{He-N2} = \frac{0.7337 \times 10^{-4}}{(p_{He} + p_{N2})} \text{ m}^2/\text{s} \tag{A.11}$$
$$D_{He-He} = \frac{1.7513 \times 10^{-4}}{(p_{He} + p_{N2})} \text{ m}^2/\text{s}. \tag{A.12}$$

The observable diffusion rate for a mixture of ^{3}He and Nitrogen gas is therefore

$$\frac{1}{D_{obs}} = \frac{p_{He}}{1.7513 \times 10^{-4} \text{ m}^2/\text{s}} + \frac{p_{N2}}{0.7337 \times 10^{-4} \text{ m}^2/\text{s}}. \tag{A.13}$$

The cell used in Helium experiments had the following pressures: $p_{He} = (0.75 \pm 0.01)$ atm and $p_{N2} = (0.10 \pm 0.01)$ atm. The theoretical estimation of the observable diffusion constant is thus $D_{obs} = (1.77 \pm 0.12) \times 10^{-4}$ m^2/s.

Appendix B

Supplement on Fourier Transforms

The Fourier Transform of $e^{-2\pi k_\circ |x|}$, where $2\pi k_\circ = 1/T_2^*$, is given by:

$$
\begin{aligned}
F\left[e^{-2\pi k_\circ |x|}\right] &= \int_{-\infty}^{\infty} e^{-2\pi k_\circ |x|} e^{-2\pi i k x} dx \\
&= \int_{-\infty}^{0} e^{-2\pi i k x} e^{2\pi k_\circ x} dx + \int_{0}^{\infty} e^{-2\pi i k x} e^{-2\pi k_\circ x} dx \\
&= \int_{-\infty}^{0} \left[\cos(2\pi k x) - i \sin(2\pi k x)\right] e^{2\pi k_\circ x} dx \\
&\quad + \int_{0}^{\infty} \left[\cos(2\pi k x) - i \sin(2\pi k x)\right] e^{-2\pi k_\circ x} dx
\end{aligned}
$$

Let $u \equiv -x$ so that $du = -dx$, then:

$$
\begin{aligned}
F\left[e^{-2\pi k_\circ |x|}\right] &= \int_{0}^{\infty} \left[\cos(2\pi k u) + i \sin(2\pi k u)\right] e^{-2\pi k_\circ u} du \\
&\quad + \int_{0}^{\infty} \left[\cos(2\pi k u) - i \sin(2\pi k u)\right] e^{-2\pi k_\circ u} du \\
&= 2 \int_{0}^{\infty} \cos(2\pi k u) e^{-2\pi k_\circ u} du \\
&= \frac{1}{\pi} \frac{k_\circ}{k^2 + k_\circ^2},
\end{aligned}
$$

which is a Lorentzian function, with: FWHM $= 2k_\circ = 1/\pi T_2^*$.

Appendix C

Imaging Parameters

The following are the descriptions of some of the most common parameters in MR imaging:

1. **Bandwidth** (BW): Anti-aliasing filter bandwidth of the receiver.

2. **Sampling Period** (Δt): Sampling period of the A/D converters.

3. **Acquisition Time or Readout Interval** ($T_{AcqTime}$): Time interval during which the signal is acquired.

4. **Field-of-View** (FOV_x, FOV_y): Image size along the x and y-coordinates.

5. **Matrix Size** ($N_x \times N_y$): Number of pixels along the readout and phase-encode directions.

6. **Spatial Resolution** (Δx, Δy): Resolution in image space.

7. **Raw Data Resolution** (Δk_x, Δk_y): Resolution in k-space.

8. **Readout Amplitude** (G_x): Amplitude of the readout gradient.

9. **Maximum Amplitude in Y-Gradient** (G_y^{max}): Maximum amplitude of y-gradient used in imaging.

10. **Incremental Amplitude in Y-Gradient** (ΔG_y): Incremental amplitude of y-gradient used in imaging.

11. **Phase Encode Interval** (t_{Gy}): Time interval during which the phase encode gradient is applied.

Below, is a set of formulas which define and connect these parameters:

$$\Delta t = 1/BW \tag{C.1}$$

$$T_{AcqTime} = \Delta t * N_x \tag{C.2}$$

$$G_y^{max} = \Delta G_y * N_y \tag{C.3}$$

$$FOV_x = 1/\Delta k_x \tag{C.4}$$

$$FOV_y = 1/\Delta k_y \tag{C.5}$$

$$\Delta x = FOV_x/N_x \tag{C.6}$$

$$\Delta y = FOV_y/N_y \tag{C.7}$$

$$\Delta k_x = \frac{\gamma}{2\pi} G_x \, \Delta t \tag{C.8}$$

$$\Delta k_y = \frac{\gamma}{2\pi} \Delta G_y \, t_{Gy} \tag{C.9}$$