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

$$J_1 = -D_{12} \nabla c_1,$$  \hspace{1cm} (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

$$D_{12} = 0.001858 \ T^{3/2} \sqrt{\frac{M_1 + M_2}{M_1 M_2}} \frac{f_D}{p \sigma_{12}^2 \Omega_D},$$  \hspace{1cm} (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; $\sigma_{12}$ is the Lennard-Jones force constant for the gas mixture, defined by $\sigma_{12} = 1/2 (\sigma_1 + \sigma_2)$; $\Omega_D$ is the collision integral.
defined by

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

where \( T^* \equiv kT/\epsilon_{12} \), \( k \) is the Boltzman gas constant, \( \epsilon_{12} = (\epsilon_1 \epsilon_2)^{1/2} \) and \( \epsilon_{12} = \sqrt{\epsilon_1 \epsilon_2} \).

Values of \( \sigma_{1(2)} \), \( \Omega_D \) and \( \epsilon_{12} \) 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: \( M_1 = M_2 = M, \epsilon_1 = \epsilon_2 \) and \( \sigma_1 = \sigma_2 \). Thus,

\[ D_{11} = 0.001858 \, T^{3/2} \sqrt{\frac{2}{M \rho \sigma_{11}^2}} \Omega_D. \quad (A.4) \]

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

\[ \frac{1}{D_{obs,1}} = \frac{1}{D_{11}(p = 1 \, atm)/ (p_1 + p_2)} + \frac{p_2}{D_{12}(p = 1 \, atm)/ (p_1 + p_2)} \]

\[ = \frac{1}{D_{11}(p = 1 \, atm)} + \frac{1}{D_{12}(p = 1 \, atm)} \]

\[ = \frac{1}{D_{11}(p = p_1)} + \frac{1}{D_{12}(p = p_2)}. \quad (A.5) \]

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:

\[
\begin{align*}
\sigma_{Xe} &= 4.047 & \epsilon_{0Xe}/k &= 231.0 & M_{Xe} &= 130.4 \\
\sigma_{N2} &= 3.798 & \epsilon_{0N2}/k &= 71.4 & M_{N2} &= 28
\end{align*}
\]

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

\[
\begin{align*}
\sigma_{Xe-N2} &= 3.9225 & \frac{\epsilon_{0Xe-N2}}{k} &= 128.42 & \frac{kT}{\epsilon_{0Xe-N2}} &= 2.398 & \Omega_D &= 1.0183 \\
\sigma_{Xe-Xe} &= 4.047 & \frac{\epsilon_{0Xe-Xe}}{k} &= 231 & \frac{kT}{\epsilon_{0Xe-Xe}} &= 1.333 & \Omega_D &= 1.2696.
\end{align*}
\]

The above parameter values yield

\[
\begin{align*}
D_{Xe-N2} &= \frac{0.1303 \times 10^{-4}}{(p_{Xe} + p_{N2})} \text{ m}^2/\text{s} & (A.7) \\
D_{Xe-Xe} &= \frac{0.0584 \times 10^{-4}}{(p_{Xe} + p_{N2})} \text{ m}^2/\text{s}. & (A.8)
\end{align*}
\]

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

\[
\frac{1}{D_{obs}} = \frac{p_{Xe}}{0.0584 \times 10^{-4} \text{ m}^2/\text{s}} + \frac{p_{N2}}{0.1303 \times 10^{-4} \text{ m}^2/\text{s}}. & (A.9)
\]

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 \( D_{obs} = (1.08 \pm 0.08) \times 10^{-5} \text{ m}^2/\text{s} \).
A.0.2 Observable Diffusion Constant for a Mixture of He-3 and Nitrogen

The relevant parameters [30] are:

\[
\begin{align*}
\sigma_{\text{He}} &= 2.551 & \epsilon_{\text{He}}/k &= 10.22 & M_{\text{He}} &= 4 \\
\sigma_{\text{N}_2} &= 3.798 & \epsilon_{\text{N}_2}/k &= 71.4 & M_{\text{N}_2} &= 28
\end{align*}
\]

At \( T = (308 \pm 10) \text{ K} \) and \( p = (p_{\text{He}} + p_{\text{N}_2}) \text{ atm} \),

\[
\begin{align*}
\sigma_{\text{He-N}_2} &= 3.1745 & \frac{\epsilon_{\text{He-N}_2}}{k} &= 27.013 & \frac{kT}{\epsilon_{\text{He-N}_2}} &= 11.587 & \Omega_D &= 0.7260 \\
\sigma_{\text{He-He}} &= 2.551 & \frac{\epsilon_{\text{He-He}}}{k} &= 10.22 & \frac{kT}{\epsilon_{\text{He-He}}} &= 30.626 & \Omega_D &= 0.6231.
\end{align*}
\] (A.10)

The above parameter values yield

\[
\begin{align*}
D_{\text{He-N}_2} &= \frac{0.7337 \times 10^{-4}}{(p_{\text{He}} + p_{\text{N}_2})} \text{ m}^2/\text{s} \quad \text{(A.11)} \\
D_{\text{He-He}} &= \frac{1.7513 \times 10^{-4}}{(p_{\text{He}} + p_{\text{N}_2})} \text{ m}^2/\text{s}. \quad \text{(A.12)}
\end{align*}
\]

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

\[
\frac{1}{D_{\text{obs}}} = \frac{p_{\text{He}}}{1.7513 \times 10^{-4} \text{ m}^2/\text{s}} + \frac{p_{\text{N}_2}}{0.7337 \times 10^{-4} \text{ m}^2/\text{s}}. \quad \text{(A.13)}
\]

The cell used in Helium experiments had the following pressures: \( p_{\text{He}} = (0.75 \pm 0.01) \text{ atm} \) and \( p_{\text{N}_2} = (0.10 \pm 0.01) \text{ atm} \). The theoretical estimation of the observable diffusion constant is thus \( D_{\text{obs}} = (1.77 \pm 0.12) \times 10^{-4} \text{ m}^2/\text{s} \).
Appendix B

Supplement on Fourier Transforms

The Fourier Transform of $e^{-2\pi k_0 |x|}$, where $2\pi k_0 = 1/T_2^*$, is given by:

\[ F[e^{-2\pi k_0 |x|}] = \int_{-\infty}^{\infty} e^{-2\pi k_0 |x|} e^{-2\pi ikx} \, dx \]
\[ = \int_{-\infty}^{0} e^{-2\pi ikx} e^{2\pi k_0 x} \, dx + \int_{0}^{\infty} e^{-2\pi ikx} e^{-2\pi k_0 x} \, dx \]
\[ = \int_{-\infty}^{0} [\cos (2\pi kx) - i \sin (2\pi kx)] e^{2\pi k_0 x} \, dx \]
\[ + \int_{0}^{\infty} [\cos (2\pi kx) - i \sin (2\pi kx)] e^{-2\pi k_0 x} \, dx \]

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

\[ F[e^{-2\pi k_0 |x|}] = \int_{0}^{\infty} [\cos (2\pi ku) + i \sin (2\pi ku)] e^{-2\pi k_0 u} \, du \]
\[ + \int_{0}^{\infty} [\cos (2\pi ku) - i \sin (2\pi ku)] e^{-2\pi k_0 u} \, du \]
\[ = 2 \int_{0}^{\infty} \cos (2\pi ku) e^{-2\pi k_0 u} \, du \]
\[ = \frac{1}{\pi k^2 + k_0^2}, \]

which is a Lorentzian function, with: FWHM = $2k_0 = 1/\pi T_2^*$.
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** ($\Delta t$): 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** ($\Delta x, \Delta y$): Resolution in image space.

7. **Raw Data Resolution** ($\Delta k_x, \Delta k_y$): 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** ($\Delta G_y$): Incremental amplitude of y-gradient used in imaging.

11. **Phase Encode Interval** ($t_{G_y}$): Time interval during which the phase encode gradient is applied.
Below, is a set of formulas which define and connect these parameters:

\[
\begin{align*}
\Delta t &= \frac{1}{BW} \quad \text{(C.1)} \\
T_{\text{AcqTime}} &= \Delta t \ast N_x \quad \text{(C.2)} \\
G_{y}^{\text{max}} &= \Delta G_y \ast N_y \quad \text{(C.3)} \\
\text{FOV}_x &= \frac{1}{\Delta k_x} \quad \text{(C.4)} \\
\text{FOV}_y &= \frac{1}{\Delta k_y} \quad \text{(C.5)} \\
\Delta x &= \text{FOV}_x / N_x \quad \text{(C.6)} \\
\Delta y &= \text{FOV}_y / N_y \quad \text{(C.7)} \\
\Delta k_x &= \frac{\gamma}{2\pi} G_x \Delta t \quad \text{(C.8)} \\
\Delta k_y &= \frac{\gamma}{2\pi} \Delta G_y tG_y \quad \text{(C.9)}
\end{align*}
\]