

ChE-402: Diffusion and Mass Transfer

Lecture 5

Intended Learning Outcome

- ✦ To solve transient problems involving both convection and diffusion.
- ✦ To further analyze origin of convection.
- ✦ To evaluate diffusion coefficients by theory as well as by using empirical models.

Transient convection and diffusion

Component 2 does not mix with liquid phase of 1

⇒ At $z = 0$, $n_2 = 0$

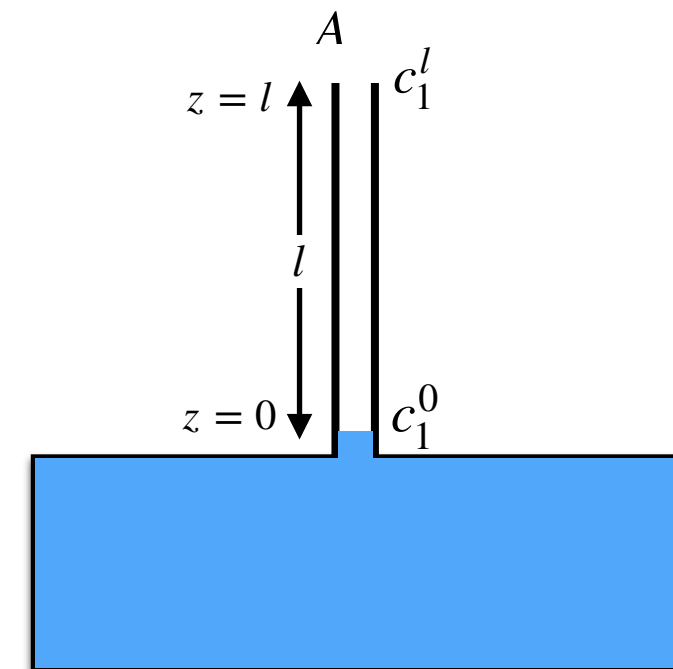
- In the steady case, we could use $v_2 = 0$ (stagnant air);
- In this unsteady case, the flux of 1 varies with time and position (for example, 1 displaces 2 immediately after $t = 0$, so $v_2 \neq 0$).

Define your system -

Define an element to do mass balance

Apply mass balance

$$\overset{o}{Accumulation} * dV = \overset{o}{Flux}|_{in} * A - \overset{o}{Flux}|_{out} * A + \overset{o}{Generation} * dV - \overset{o}{Consumption} * dV$$



Fast evaporation by
diffusion and
convection

Transient convection and diffusion

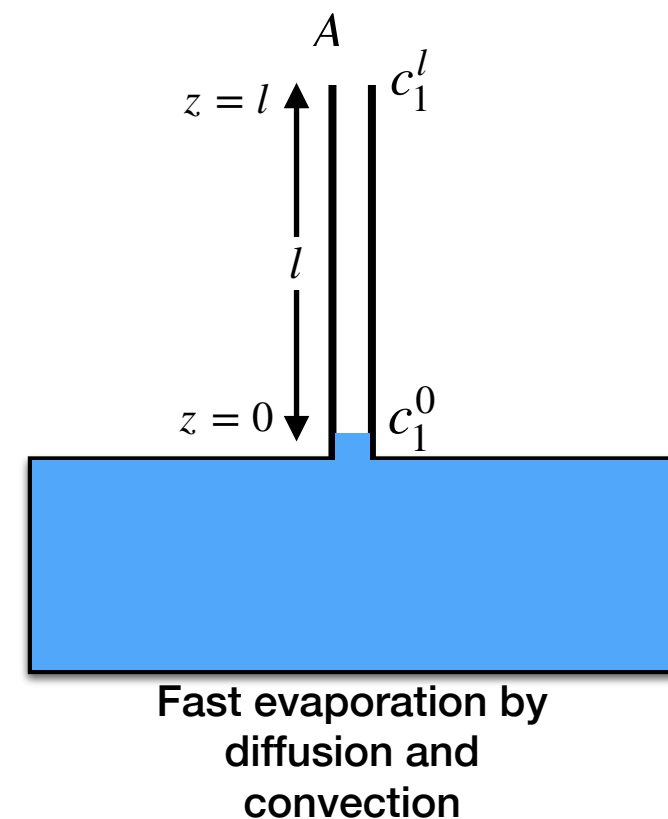
$$\frac{\partial c_1}{\partial t} = - \frac{\partial n_1}{\partial z}$$

$$v^v = c_1 \bar{V}_1 v_1 + c_2 \bar{V}_2 v_2 = \bar{V}_1 n_1 + \bar{V}_2 n_2$$

$$\frac{\partial c_1}{\partial t} = D \frac{\partial^2 c_1}{\partial z^2} - \frac{\partial}{\partial z}(c_1 v^v)$$

Apply mass balance for component 2

$$\overset{o}{Accumulation} * dV = \overset{o}{Flux} |_{in} * A - \overset{o}{Flux} |_{out} * A + \overset{o}{Generation} * dV - \overset{o}{Consumption} * dV$$



Transient convection and diffusion

$$\frac{\partial c_1}{\partial t} = - \frac{\partial n_1}{\partial z}$$

$$\frac{\partial c_2}{\partial t} = - \frac{\partial n_2}{\partial z}$$

$$v^v = c_1 \bar{V}_1 v_1 + c_2 \bar{V}_2 v_2 = \bar{V}_1 n_1 + \bar{V}_2 n_2$$

Multiply each term with molar volume and add

$$\frac{\partial}{\partial t}(\bar{V}_1 c_1 + \bar{V}_2 c_2) = - \frac{\partial}{\partial z}(\bar{V}_1 n_1 + \bar{V}_2 n_2) = - \frac{\partial v^v}{\partial z}$$

$$\bar{V}_1 c_1 + \bar{V}_2 c_2 \approx y_1 + y_2 = 1 \quad \text{We are dealing with vapor phase}$$

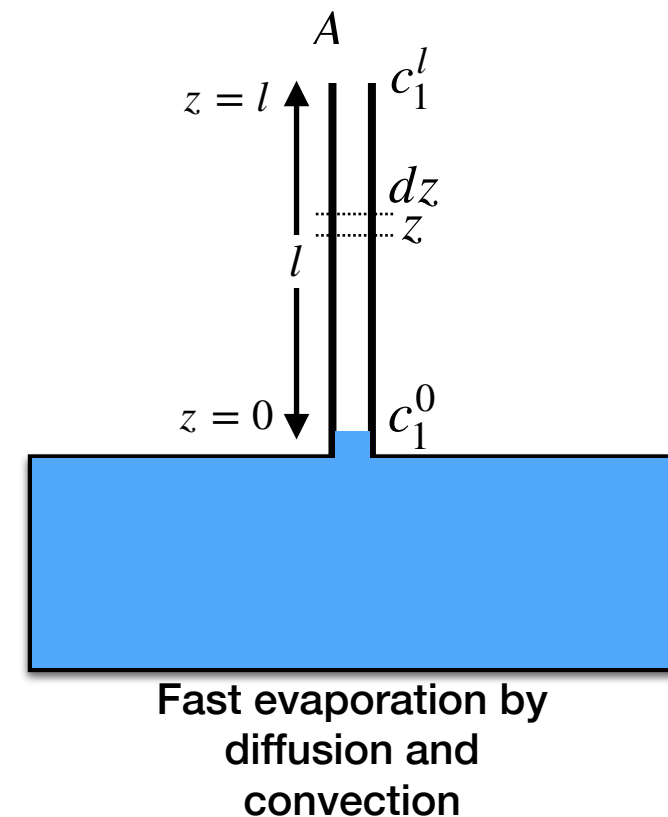
Implications $\frac{\partial}{\partial t}(\bar{V}_1 c_1 + \bar{V}_2 c_2) = 0 = - \frac{\partial v^v}{\partial z}$

- Volume average velocity, v^v , is constant, and does not change with z

$$v^v = \bar{V}_1 n_1 + \bar{V}_2 n_2 = \bar{V}_1 n_1 |_{z=0}$$

- At $z = 0$, $n_2 = 0$

$$\Rightarrow n_1 |_{z=0} = -D \frac{\partial c_1}{\partial z} |_{z=0} + c_1 v^v |_{z=0} = -D \frac{\partial c_1}{\partial z} |_{z=0} + c_1 \bar{V}_1 n_1 |_{z=0}$$



Transient convection and diffusion

$$n_1|_{z=0} = -D \frac{\partial c_1}{\partial z} \Big|_{z=0} + c_1 \bar{V}_1 n_1|_{z=0} \quad \Rightarrow \quad n_1|_{z=0} = \left(\frac{-D \frac{\partial c_1}{\partial z}}{1 - c_1 \bar{V}_1} \right) \Big|_{z=0}$$

Constant $\nearrow v^v = \bar{V}_1 n_1 + \bar{V}_2 n_2 = \bar{V}_1 n_1|_{z=0}$

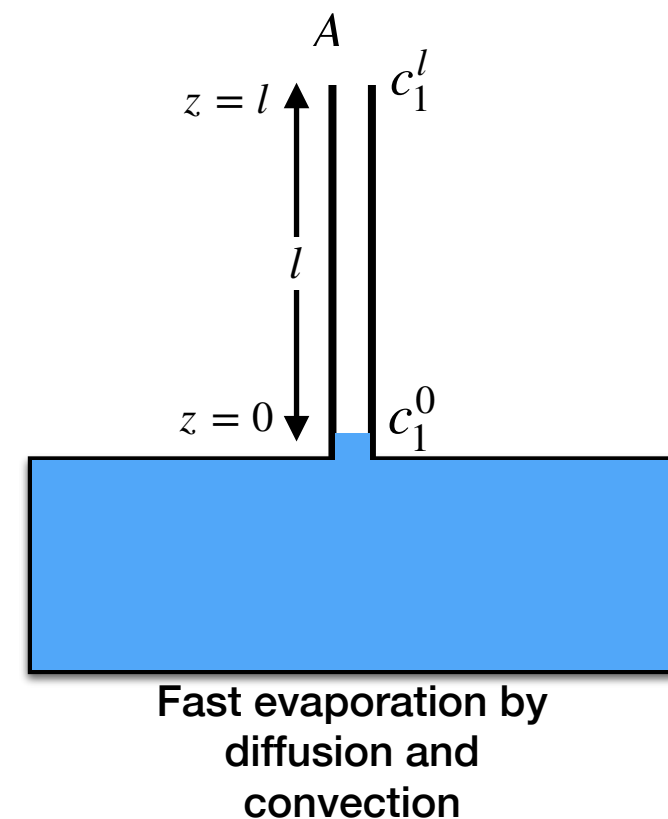
$$\frac{\partial c_1}{\partial t} = D \frac{\partial^2 c_1}{\partial z^2} - \frac{\partial}{\partial z}(c_1 v^v)$$

$$\Rightarrow \frac{\partial c_1}{\partial t} = D \frac{\partial^2 c_1}{\partial z^2} - v^v \frac{\partial c_1}{\partial z}$$

$$\Rightarrow \frac{\partial c_1}{\partial t} = D \frac{\partial^2 c_1}{\partial z^2} - \bar{V}_1 n_1|_{z=0} \frac{\partial c_1}{\partial z}$$

$$\Rightarrow \frac{\partial c_1}{\partial t} = D \frac{\partial^2 c_1}{\partial z^2} - \bar{V}_1 \left(\frac{-D \frac{\partial c_1}{\partial z}}{1 - c_1 \bar{V}_1} \right) \Big|_{z=0} \frac{\partial c_1}{\partial z}$$

$$\Rightarrow \frac{\partial c_1}{\partial t} = D \frac{\partial^2 c_1}{\partial z^2} + D \left(\frac{\bar{V}_1 \frac{\partial c_1}{\partial z}}{1 - c_1 \bar{V}_1} \right) \Big|_{z=0} \frac{\partial c_1}{\partial z}$$



Transient convection and diffusion

$$\frac{\partial c_1}{\partial t} = D \frac{\partial^2 c_1}{\partial z^2} + D \left(\frac{\bar{V}_1 \frac{\partial c_1}{\partial z}}{1 - c_1 \bar{V}_1} \right) \Big|_{z=0} \frac{\partial c_1}{\partial z}$$

$$\zeta = \frac{z}{\sqrt{4Dt}}$$

Initial condition: $t = 0, c_1 = 0$

Boundary conditions

$$t > 0 \quad c_1 \Big|_{z=0} = c_1^{sat}$$

$$\frac{d^2 c_1}{d\zeta^2} + 2(\zeta - \phi) \frac{dc_1}{d\zeta} = 0$$

$$\phi = -\frac{1}{2} \left(\frac{\bar{V}_1 \frac{\partial c_1}{\partial \zeta}}{1 - c_1 \bar{V}_1} \right) \Big|_{z=0}$$

Dimensionless velocity, which determines extent of diffusion-led convection and the movement of the interface ($z = 0$).

- Higher diffusive flux leads to convection.
- Higher concentration leads to convection.

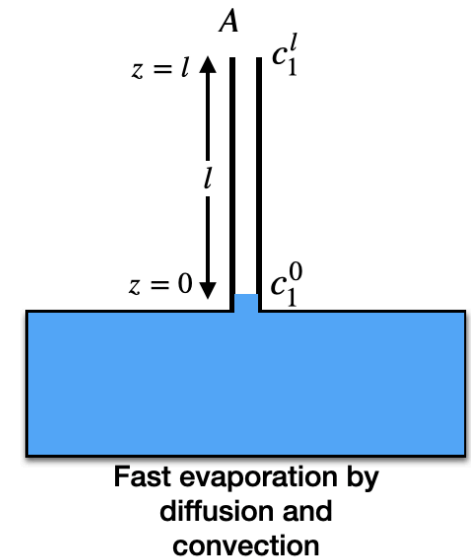
Boundary/Initial conditions:

$$c_1 \Big|_{\zeta=0} = c_1^{sat}$$

$$c_1 \Big|_{\zeta=\infty} = 0$$

$$\frac{c_1}{c_1^{sat}} = \frac{1 - \operatorname{erf}(\zeta - \phi)}{1 + \operatorname{erf} \phi}$$

$$\bar{V}_1 c_1^{sat} = \left(1 + \frac{1}{\sqrt{\pi}(1 + \operatorname{erf} \phi)\phi e^{\phi^2}} \right)^{-1}$$



When there is no convection

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial z^2}$$

$$\frac{d^2 c_1}{d\zeta^2} + 2\zeta \frac{dc_1}{d\zeta} = 0$$

$$\frac{c(z, t) - c_S}{c_\infty - c_S} = \operatorname{erf} \zeta$$

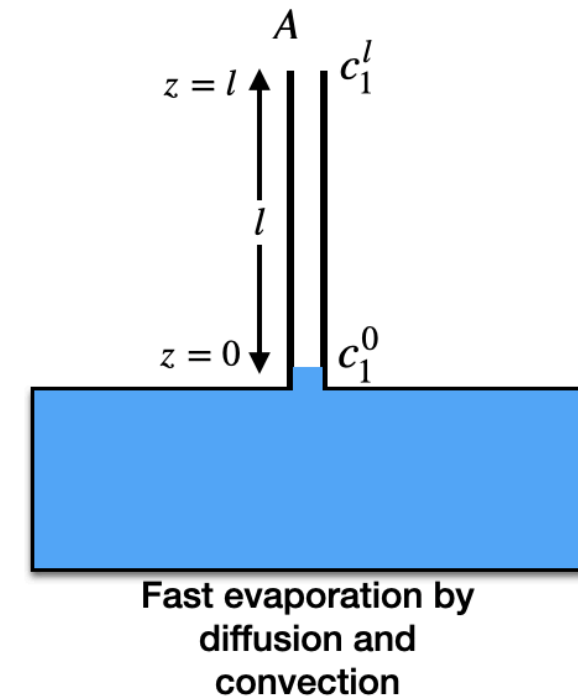
Summary: transient convection and diffusion

$$\phi = -\frac{1}{2} \left(\frac{\bar{V}_1 \frac{\partial c_1}{\partial \zeta}}{1 - c_1 \bar{V}_1} \right) \Big|_{z=0}$$

$$\bar{V}_1 c_1^{\text{sat}} = \left(1 + \frac{1}{\sqrt{\pi} (1 + \operatorname{erf} \phi) \phi e^{\phi^2}} \right)^{-1}$$

$$\zeta = \frac{z}{\sqrt{4Dt}}$$

$$\frac{c_1}{c_1^{\text{sat}}} = \frac{1 - \operatorname{erf} (\zeta - \phi)}{1 + \operatorname{erf} \phi}$$

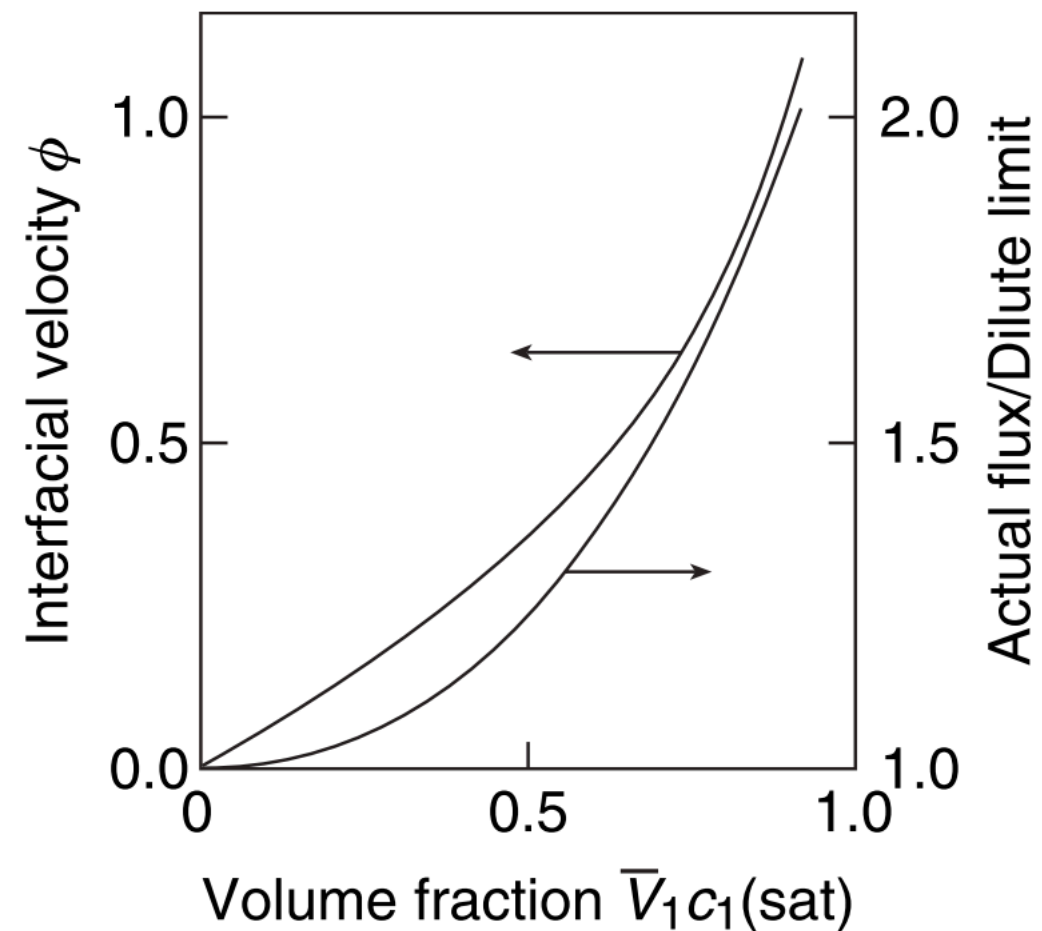


$$n_1 \Big|_{z=0} = \left(\frac{-D \frac{\partial c_1}{\partial z}}{1 - c_1 \bar{V}_1} \right) \Big|_{z=0} = \sqrt{\frac{D}{\pi t}} \left(\frac{1}{1 - \bar{V}_1 c_1^{\text{sat}}} \right) \frac{e^{-\phi^2}}{1 + \operatorname{erf} \phi} c_1^{\text{sat}}$$

Summary: transient convection and diffusion

$$\phi = -\frac{1}{2} \left(\frac{\bar{V}_1 \frac{\partial c_1}{\partial \zeta}}{1 - c_1 \bar{V}_1} \right) \Big|_{z=0}$$

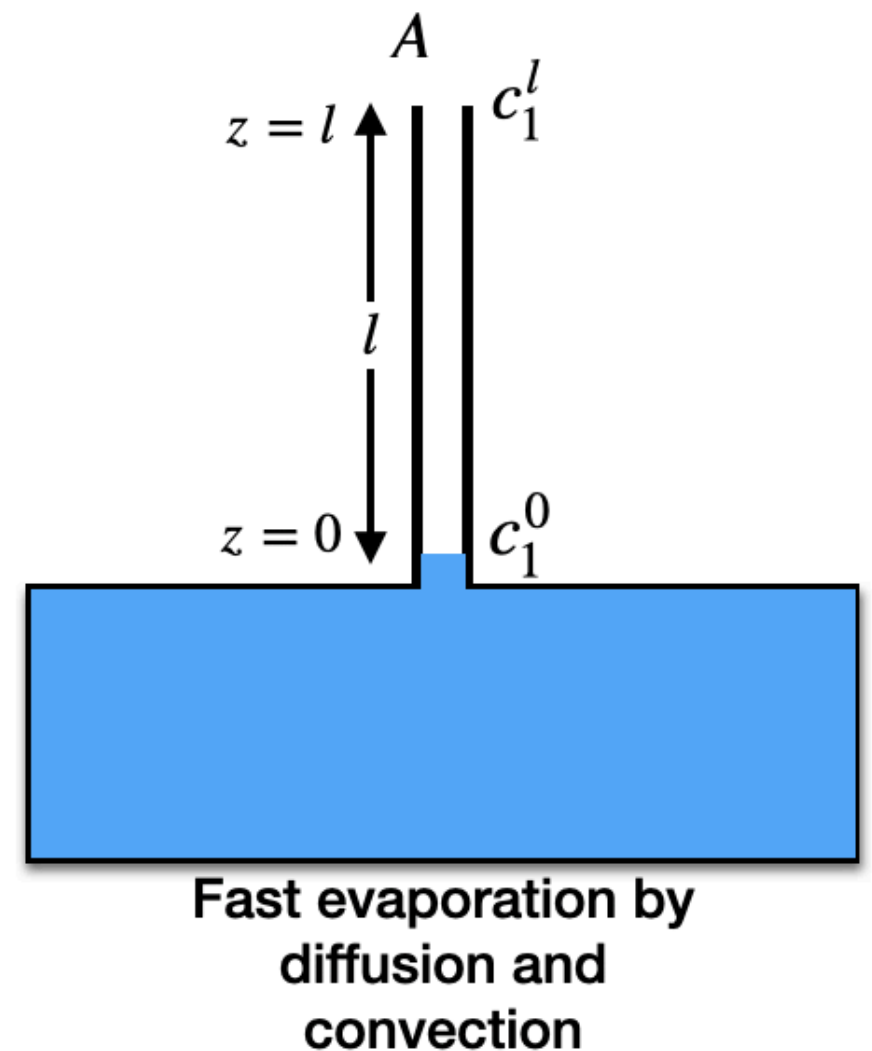
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In which of the following cases, convection can not be neglected

- a) Diffusive flux at $z = 0$ is high
- b) y_1 at $z = 0$ is 0.05
- c) y_1 at $z = L$ is 0.5

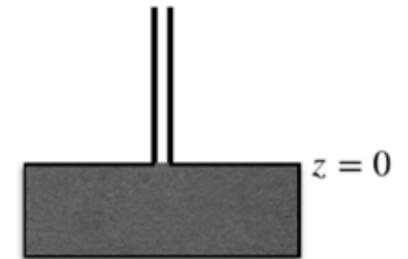
$$\phi = -\frac{1}{2} \left(\frac{\bar{V}_1 \frac{\partial c_1}{\partial \zeta}}{1 - c_1 \bar{V}_1} \right) \Big|_{z=0}$$



In-class exercise problem

A container filled with liquid aniline (molar mass: 93.13 g/mol) is stored in a large room. The container has a cylindrical tube with length of 10 cm and a diameter of 1 cm. Initially, the cylindrical tube is filled with air (pressure of 1 atm), and aniline is prevented from evaporation. At time $t = 0$, the evaporation of aniline is allowed.

- Saturation vapor pressure of aniline: 10 kPa
- The diffusion coefficient of the aniline vapor in air: $9 \times 10^{-2} \text{ cm}^2/\text{s}$.
- Aniline vapor can be treated as ideal gas.
- Assume a concentration of 0 at $z = 10 \text{ cm}$, and saturation conditions at $z = 0$.

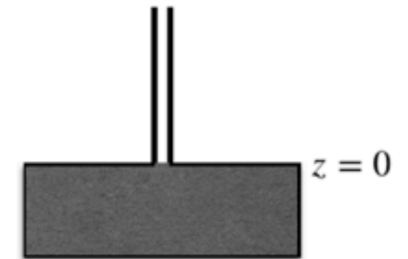


1. Neglecting convection, calculate the partial pressure of aniline vapor at $z = 1.5 \text{ cm}$ after 25 seconds.
2. Estimate the error in neglecting convection.

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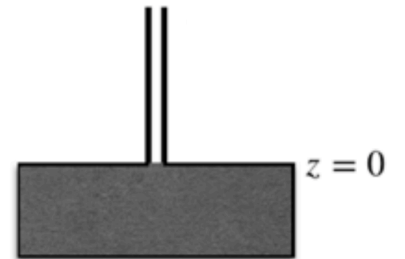
1. Neglecting convection, calculate the partial pressure of aniline vapor at $z = 1.5 \text{ cm}$ after 25 seconds.
2. Estimate the error in neglecting convection.

When convection is neglected;

In-class exercise problem

When convection is considered

$$\bar{V}_1 c_1^{\text{sat}} = \left(1 + \frac{1}{\sqrt{\pi}(1 + \operatorname{erf} \phi)\phi e^{\phi^2}} \right)^{-1}$$



In-class exercise problem

$$\frac{c_1}{c_1^{\text{sat}}} = \frac{1 - \operatorname{erf}(\zeta - \phi)}{1 + \operatorname{erf} \phi}$$

Diffusion coefficients in gases, liquids and solids

Understanding the driving force for diffusion

- Diffusion flux is essentially a flow driven by force (the gradient of chemical potential).
- If there is a flow, there should be frictional force opposing the flow.

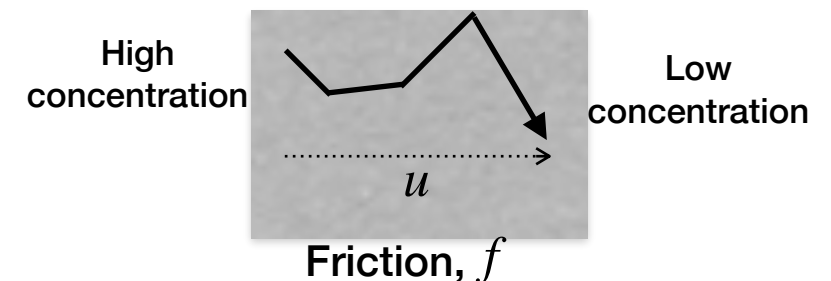
Frictional drag = chemical potential force

$$fu = -\frac{d\mu}{dz} \quad \Rightarrow \quad u = -\frac{1}{f} \frac{d\mu}{dz}$$

$$\text{flux} = uc = -\frac{c}{f} \frac{d\mu}{dz} = -\left[\frac{k_B T}{f} \frac{d \ln(\hat{f}/P)}{d \ln c} \right] \frac{dc}{dz} = -D \frac{dc}{dz}$$

$$D = \frac{k_B T}{f} \frac{d \ln(\hat{f}/P)}{d \ln c} = D_o \frac{d \ln(\hat{f}/P)}{d \ln c}$$

$$D_o = \frac{k_B T}{f}$$



Diffusivity is inversely proportional to frictional force

Stoke's Einstein Equation

EPFL

Comparison of diffusion coefficients

$$D_o = \frac{RT}{f}$$

f_{gas}

f_{liquid}

f_{solid}

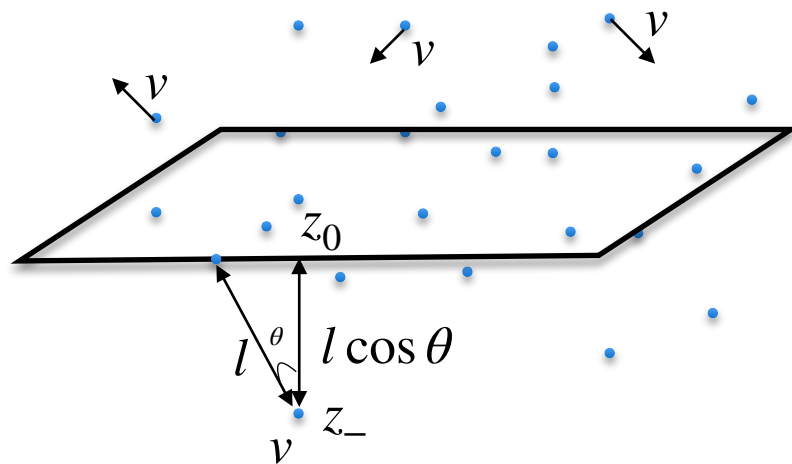
D_{gas}

D_{liquid}

D_{solid}

Diffusion coefficient of gas

l = mean free path



$$z_0 = z_- + l \cos \theta$$

$$\frac{dn}{dt} \uparrow = c|_{z_-} v_z \quad v_z = v \cos \theta$$

Velocity of particles are thermally distributed

$$g(v) = \left(\frac{m}{2\pi k_B T} \right)^{3/2} e^{-\frac{mv^2}{2k_B T}}$$

$$\left\langle \frac{dn}{dt} \uparrow \right\rangle = \int c|_{z_-} v_z g(v) d^3v = \int_{-\infty}^{\infty} dv_x \int_{-\infty}^{\infty} dv_y \int_0^{\infty} c|_{z_-} v_z g(v) dv_z$$

$$J|_{z=z_0} = \left\langle \frac{dn}{dt} \uparrow \right\rangle - \left\langle \frac{dn}{dt} \downarrow \right\rangle = -D \frac{dc}{dz} \Big|_{z=z_0}$$

Employ Taylor series for $c|_{z_-}$ $f(x) = f(x_0) + (x - x_0) \frac{df}{dx} \Big|_{x=x_0}$

$$c|_{z_-} = c|_{z_0} + (z_- - z_0) \frac{dc}{dz} \Big|_{z=z_0}$$

$$c|_{z_-} = c|_{z_0} - l \cos \theta \frac{dc}{dz} \Big|_{z=z_0}$$

Diffusion coefficient of gas

$$J|_{z=z_0} = \left\langle \frac{dn}{dt} \uparrow \right\rangle - \left\langle \frac{dn}{dt} \downarrow \right\rangle$$

$$J|_{z=z_0} = \int_{-\infty}^{\infty} dv_x \int_{-\infty}^{\infty} dv_y \int_{-\infty}^{\infty} \left(c|_{z_-} v_z - c|_{z_+} v_z \right) g(v) dv_z$$

$$c|_{z_-} v_z = c|_{z_0} v_z - l \cos \theta \frac{dc}{dz} \Big|_{z=z_0} v_z$$

$$c|_{z_+} v_z = c|_{z_0} v_z + l \cos \theta \frac{dc}{dz} \Big|_{z=z_0} v_z$$

$$\Rightarrow J|_{z=z_0} = - \int_{-\infty}^{\infty} dv_x \int_{-\infty}^{\infty} dv_y \int_{-\infty}^{\infty} \left(2l \cos \theta \frac{dc}{dz} \Big|_{z=z_0} v_z \right) g(v) dv_z$$

$$\Rightarrow J|_{z=z_0} = - 2l \frac{dc}{dz} \Big|_{z=z_0} \left(\int_{-\infty}^{\infty} dv_x \int_{-\infty}^{\infty} dv_y \int_{-\infty}^{\infty} \cos \theta v_z g(v) dv_z \right) = - D \frac{dc}{dz} \Big|_{z=z_0}$$

$\frac{\bar{v}}{6}$

$\bar{v} = \sqrt{\frac{8k_B T}{\pi m}}$

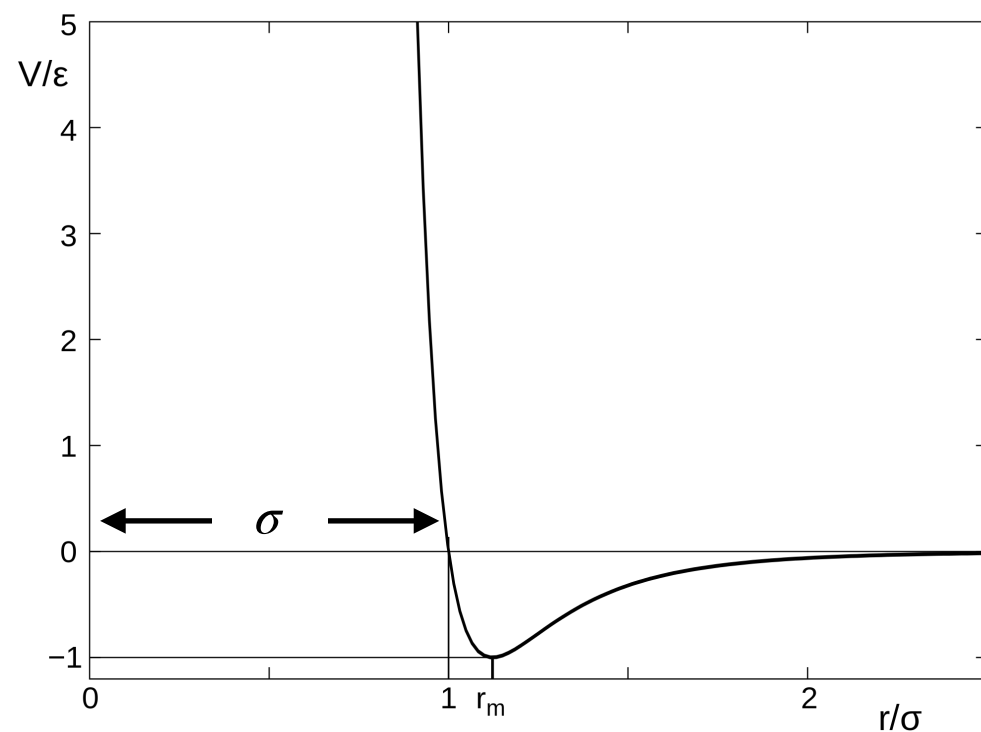
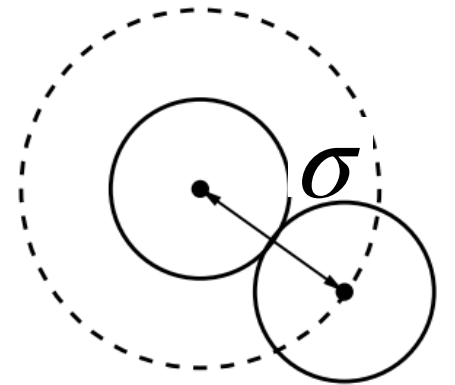
$$\Rightarrow J|_{z=z_0} = - 2l \frac{dc}{dz} \Big|_{z=z_0} \left(\frac{\bar{v}}{6} \right) = - D \frac{dc}{dz} \Big|_{z=z_0}$$

$$D = \frac{1}{3} \bar{v} l$$

Diffusion coefficient of gas

$$D = \frac{1}{3} \bar{v} l$$

$$\bar{v} = \sqrt{\frac{8k_B T}{\pi m}}$$



$$V_{LJ} = 4\epsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right]$$

$$l = \frac{k_B T / P}{\left(\frac{\pi}{4} \sigma^2 \right)} = \frac{\text{volume of space per molecule}}{\text{cross-sectional area}}$$

Mean free path is much bigger than average distance between gas molecules

$$D = \frac{1}{3} \bar{v} l = \frac{8}{3} \sqrt{\frac{2}{m}} \left(\frac{k_B T}{\pi} \right)^{3/2} \frac{1}{P \sigma^2}$$

Binary diffusion coefficient for gas

Chapman-Enskog theory: Empirically derived (accurate to 8%)

$$D_{12} = \frac{1.86 * 10^{-3} * T^{1.5} * (1/M_1 + 1/M_2)^{0.5}}{P\sigma_{12}^2\Omega}$$

Ω = Collision Integral

D in cm²/s

T in Kelvin

P in atm

M in g/mole

σ_{12} in Angstrom

$$\sigma_{12} = \frac{\sigma_1 + \sigma_2}{2}$$

$$\epsilon_{12} = \sqrt{\epsilon_1\epsilon_2}$$

$$V_{LJ} = 4\epsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right]$$

Table 5.1-2 Lennard-Jones potential parameters found from viscosities

Substance		$\sigma(\text{\AA})$	$\epsilon_{12}/k_B(\text{K})$
Ar	Argon	3.542	93.3
He	Helium	2.551	10.2
Kr	Krypton	3.655	178.9
Ne	Neon	2.820	32.8
Xe	Xenon	4.047	231.0
Air	Air	3.711	78.6
Br ₂	Bromine	4.296	507.9
CCl ₄	Carbon tetrachloride	5.947	322.7
CHCl ₃	Chloroform	5.389	340.2
CH ₂ Cl ₂	Methylene chloride	4.898	356.3
CH ₃ Cl	Methyl chloride	4.182	350.0
CH ₃ OH	Methanol	3.626	481.8
CH ₄	Methane	3.758	148.6
CO	Carbon monoxide	3.690	91.7
CO ₂	Carbon dioxide	3.941	195.2

Table 5.1-3 The collision integral Ω

$k_B T/\epsilon_{12}$	Ω	$k_B T/\epsilon_{12}$	Ω	$k_B T/\epsilon_{12}$	Ω
0.30	2.662	1.65	1.153	4.0	0.8836
0.40	2.318	1.75	1.128	4.2	0.8740
0.50	2.066	1.85	1.105	4.4	0.8652
0.60	1.877	1.95	1.084	4.6	0.8568
0.70	1.729	2.1	1.057	4.8	0.8492
0.80	1.612	2.3	1.026	5.0	0.8422
0.90	1.517	2.5	0.9996	7	0.7896
1.00	1.439	2.7	0.9770	9	0.7556
1.10	1.375	2.9	0.9576	20	0.6640
1.30	1.273	3.3	0.9256	60	0.5596
1.50	1.198	3.7	0.8998	100	0.5130
1.60	1.167	3.9	0.8888	300	0.4360

Source: Data from Hirschfelder *et al.* (1954).

Diffusion at high pressure

$$D = \frac{8}{3} \sqrt{\frac{2}{m}} \left(\frac{k_B T}{\pi} \right)^{3/2} \frac{1}{P \sigma^2}$$

$$DP = \text{constant}$$

$$D_1 P_1 = D_2 P_2$$

$$D_2 = D_1 \frac{P_1}{P_2}$$

Diffusion coefficient in liquid

One can primarily use the Stoke's-Einstein equation

$$D_o = \frac{k_B T}{f} = \frac{k_B T}{6\pi\eta R}$$

Stoke's law $f = 6\pi\eta R$

Valid when $\frac{R_{solute}}{R_{solvent}} \geq 5$

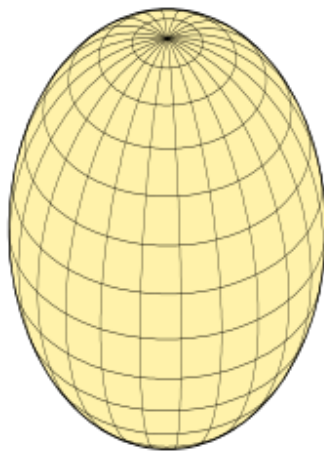
Shape effect

Spherical



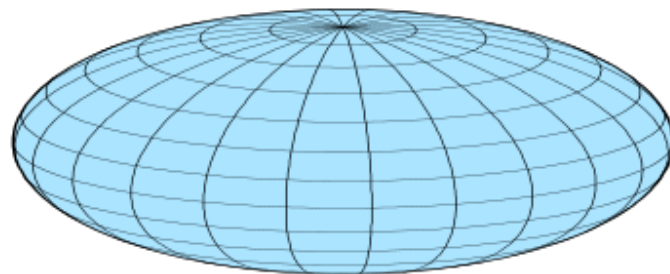
$$D_o = \frac{k_B T}{f} = \frac{k_B T}{6\pi\eta R}$$

Prolate ellipsoid



$$D_o = \frac{k_B T}{f} = \frac{k_B T}{6\pi\eta \left[\frac{(a^2 - b^2)^{1/2}}{\ln \left(\frac{a + (a^2 - b^2)^{1/2}}{b} \right)} \right]}$$

Oblate ellipsoid



$$D_o = \frac{k_B T}{f} = \frac{k_B T}{6\pi\eta \left[\frac{(a^2 - b^2)^{1/2}}{\tan^{-1} \left(\frac{(a^2 - b^2)^{1/2}}{b} \right)} \right]}$$

Diffusion coefficient in liquid

When $R_{solute} \approx R_{solvent}$

$$D = \frac{7.8 \cdot 10^{-8} T \sqrt{\phi M_2}}{\eta \bar{V}_1^{-0.6}}$$

Wilke-Chang correlation

D is diffusion coefficient of solute in cm²/s

ϕ is empirical (1 for most organic solvents, 1.5 for alcohols, 2.6 for water)

$\bar{V}_1$ is molar volume of solute in cm³/mol

M_2 is molecular weight of solvent in daltons

η is viscosity in centipoise

Diffusion coefficients in solids

Table 5.3-1 *Diffusion coefficients at 25 °C in some characteristic solids*

Solid	Solute	D (cm ² /sec)
Iron (α Fe; BCC)	Fe	$3 \cdot 10^{-48}$
	C	$6 \cdot 10^{-21}$
	H ₂	$2 \cdot 10^{-9}$
Iron (α Fe; FCC)	Fe	$8 \cdot 10^{-55}$
	C	$3 \cdot 10^{-31}$
Copper	Cu	$8 \cdot 10^{-42}$
	Zn	$2 \cdot 10^{-38}$

Diffusion is activated

$$D = D_o e^{-\frac{\Delta E}{RT}}$$

Extremely small diffusion coefficient

- 1) Almost all transport takes place through defects in the solid, especially along grain-boundaries.
- 2) Transport approaches the limit of semi-infinite media rather than the thin film.
- 3) Diffusion of hydrogen in solids is an exception because hydrogen dissociates in atomic hydrogen. The electrons of hydrogen disperse into the metallic electron cloud. The proton being much smaller than inter-atomic distances can reside at the inter-metallic interstitial positions and diffuse relatively fast through the lattice.

Exercise problem 1: calculate mean-free path, mean velocity and D of helium at 1 atm and 25 °C.

$$l = \frac{k_B T / P}{\left(\frac{\pi}{4} \sigma^2\right)} = \frac{\text{volume occupied by single molecule}}{\text{cross-sectional area}}$$

$$\bar{v} = \sqrt{\frac{8k_B T}{\pi m}}$$

$$D = \frac{1}{3} \bar{v} l = \frac{8}{3} \sqrt{\frac{2}{m}} \left(\frac{k_B T}{\pi}\right)^{3/2} \frac{1}{P \sigma^2}$$

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Exercise problem 2: In previous problem, calculate how fast a gas molecule is colliding. Also, calculate average distance between molecules and compare to mean free path.

Exercise problem 3: Calculate D for helium in argon at 1 atm and 25 °C with the Chapman-Enskog theory

$$D_{12} = \frac{1.86 * 10^{-3} * T^{1.5} * (1/M_1 + 1/M_2)^{0.5}}{P \sigma_{12}^2 \Omega}$$

D in cm²/s
 T in Kelvin
 P in atm
 M in g/mole
 σ_{12} in Angstrom

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He	Helium	2.551	10.2

Table 5.1-3 The collision integral Ω

$k_B T/\varepsilon_{12}$	Ω	$k_B T/\varepsilon_{12}$	Ω	$k_B T/\varepsilon_{12}$	Ω
0.30	2.662	1.65	1.153	4.0	0.8836
0.40	2.318	1.75	1.128	4.2	0.8740
0.50	2.066	1.85	1.105	4.4	0.8652
0.60	1.877	1.95	1.084	4.6	0.8568
0.70	1.729	2.1	1.057	4.8	0.8492
0.80	1.612	2.3	1.026	5.0	0.8422
0.90	1.517	2.5	0.9996	7	0.7896
1.00	1.439	2.7	0.9770	9	0.7556
1.10	1.375	2.9	0.9576	20	0.6640
1.30	1.273	3.3	0.9256	60	0.5596
1.50	1.198	3.7	0.8998	100	0.5130
1.60	1.167	3.9	0.8888	300	0.4360

Source: Data from Hirschfelder *et al.* (1954).