Supplemental material to

Spinning drop dynamics in miscible and immiscible environments

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ROTATIONAL LENGTH FOR SPINNING WATER-GLYCEROL MIXTURES

In order to discuss the effect of diffusion on our experiments, we need to start by understanding whether there is an influence of the centripetal forcing on diffusion itself, i.e. if we can consider diffusion to be Fickian. We can indeed expect, in principle, the concentration profile of fluids in the capillary to be set by the rotation, in analogy with the exponential profile set by gravity [1]. This characteristic length scale of the problem determines then the effect of the centripetal forcing on diffusion: if this "rotational length" l_ω is much bigger than the capillary diameter d_c, we can consider the system to be freely diffusing without any influence of the centrifugal forcing, and diffusion to be Fickian. With a procedure analogous to that described in Ref. [1], at equilibrium we can write the variation of chemical potential along the radial coordinate r as

$$\left(\frac{d\mu_i}{dr}\right)_T = M_i\omega^2 r \tag{1}$$

for each species i, whose molecular weight is M_i. Similarly, for the pressure variation

$$\frac{dp}{dr} = \rho(r)\omega^2 r \tag{2}$$

where $\rho(r)$ is the density at distance r from the axis of rotation. If we consider the system as an ideal mixture we have then from thermodynamics

$$d\mu_i = v_i dp + \sum_j \frac{\partial \mu_i}{\partial x_j} dx_j \tag{3}$$

and

$$\mu_i = \mu_i^0 + RT\ln x_i \tag{4}$$

with T being the temperature, R the universal gas constant, v_i and x_i the partial molar volume and mole fraction of species i. Combining eq. 1 to 4 we get

$$\frac{RT}{x_i}\frac{dx_i}{dr} = \left[M_i - \rho(r)v_i\right]\omega^2 r \tag{5}$$

If we approximate $\rho(r)$ with a constant equal to the average density of the two fluids, i.e. $\rho(r) = \rho_{av}$, Eq. 5 can be integrated analytically. This approximation works well for fluids that have a similar density (in our case $\rho_{H_2O} = 1000\,\mathrm{kg/m^3}$ and $\rho_{gly} = 1260\,\mathrm{kg/m^3}$) and allows us to get an estimate of the rotational length of our system [1]. Integration of eq. 5 yields

$$x_i\left(r\right) \simeq x_i\left(0\right)\exp\left\{-\frac{(\rho_{av}v_i - M_i)\omega^2}{2RT}r^2\right\} \tag{6}$$

from which we can define the square rotational length l_ω^2,

$$l_\omega^2 = \frac{2RT}{(\rho_{av}v_i - M_i)\omega^2}\,. \tag{7}$$

For the case of water in glycerol at 15000 rpm, we obtain

$$l_\omega = \frac{1}{\omega}\sqrt{\frac{2RT}{\rho_{av}v_w - M_w}} \approx 2.75\,\mathrm{m} \gg d_c \tag{8}$$

and we can thus assume diffusion not to be affected by the centripetal forcing, and thus to be described by Fick's law.

[1] O. Obidi, A. H. Muggeridge, and V. Vesovic, Physical Review E **95** (2017), ISSN 2470-0045, 2470-0053, URL `https://link.aps.org/doi/10.1103/PhysRevE.95.022138`.