Mixed-Mode Instability in a Ternary Mixture

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This is a summary of the experimental setup from [1] for getting a nice diffusion-driven gravitational instability in a simple ternary mixture: a solution of sugar on top of a solution of salt (in the beginning the density should be higher on the bottom so it starts off stable). I suggest our target to be to reproduce some aspects (at least the pretty picture) in Fig. 4 of [1] – this is the mixed-mode instability. This is when one starts with an unstable (heavy on top of light fluid) situation but the differential diffusion effects also affect the instability. One can also consider the simpler DLC instability (we did this in the PRE for compressible flow) or the classical salt-fingering DD instability, but the mixed-mode one seems most interesting and complex.

We consider a ternary mixture of salt (KCl, species 1, molar mass $74.55\,g{\cdot}mol^{-1}$, denoted by A in paper), sugar (sucrose, species 2, molar mass $342.3\,g{\cdot}mol^{-1}$, denoted by B in paper) and water (species 3, molar mass $18.02\,g{\cdot}mol^{-1}$). The initial configuration is salt solution on top of sugar solution.

Our equation of state is defined as

$$\sum_i \frac{\rho_i}{\bar{\rho}_i} = \rho \sum_i \frac{w_i}{\bar{\rho}_i} = 1, \tag{1}$$

where $w_i = \rho_i/\rho$ is the mass fraction and $\bar{\rho}_i$ are the (potentially ficticious) pure component densities. We can write this in the form

$$\frac{\rho - \rho_1 - \rho_2}{\bar{\rho}_3} + \frac{\rho_1}{\bar{\rho}_1} + \frac{\rho_2}{\bar{\rho}_2} = 1. \tag{2}$$

In the paper, the approximate formula for the density is

$$\rho = \rho_0 \left(1 + \alpha_1 Z_1 + \alpha_2 Z_2\right) = \rho_0 \left(1 + \frac{\alpha_1}{M_1}\rho_1 + \frac{\alpha_2}{M_2}\rho_2\right), \tag{3}$$

where $\rho_0 = \bar{\rho}_3 = 10^3\,g/l$ (I use liter here since they use it in the paper, we can use cm^3 in the actual runs) is the density of water, α_k are constants and Z_k is the number of moles of each component, related to the partial density via $\rho_k = Z_k M_k$, where M_k is the molar mass. By comparing (2) and (3) we get

$$1 - \frac{\rho_0}{\bar{\rho}_k} = \rho_0 \frac{\alpha_k}{M_k},$$

which now gives us the value of

$$\bar{\rho}_1 = 2.81 \text{ and } \bar{\rho}_2 = 1.55$$

from the values of α tabulated in table I of the paper, $\alpha_1 = 4.8 \cdot 10^{-2}\,l/mol$ for KCl and $\alpha_2 = 12.2 \cdot 10^{-2}\,l/mol$ for sucrose. BTW, when used for glycerol ($\alpha = 2.3$ from Table I and molar mass 92.2), this gives $\bar{\rho} = 1.33$, which is close to the correct value of 1.26, as it should since glycerol exists as an actual liquid pure phase.
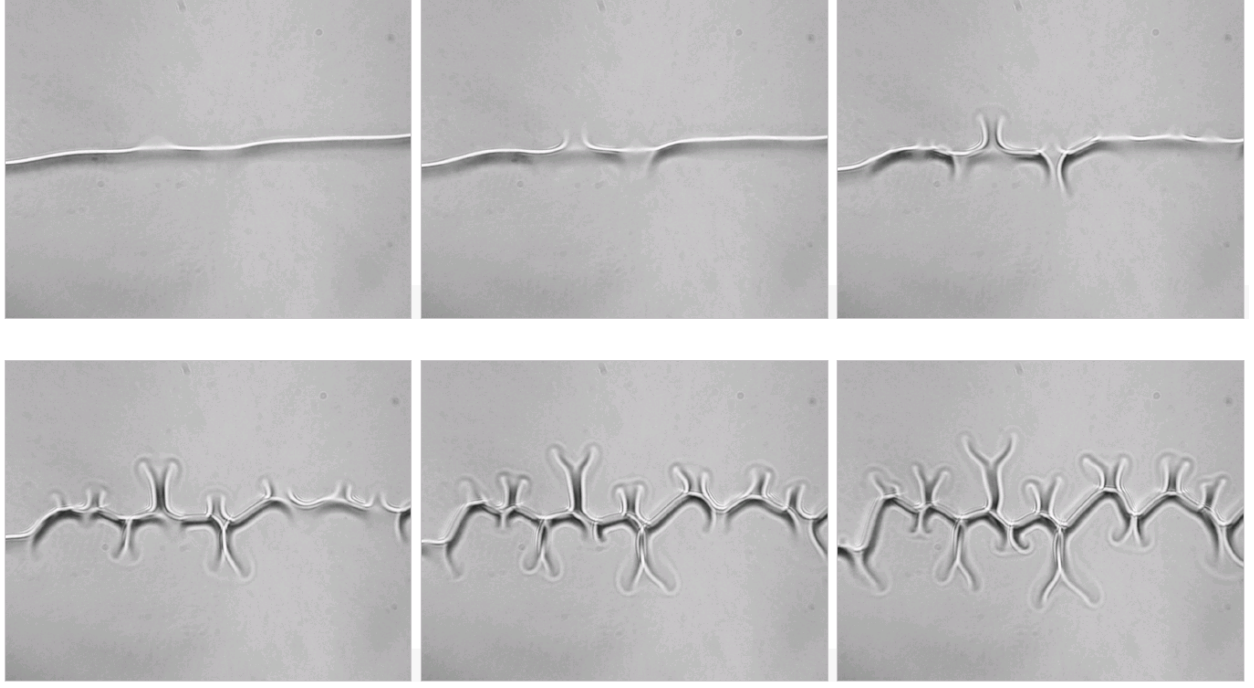
Figure 1: MMI

For the diffusion coefficients, these mixtures are at what can be considered "infinite dilution" so there is essentially no coupling between the different components (from this pespective it is a bit boring). In this limit the approximation proposed in [2] suggest setting the Maxwell-Stefan diffusion coefficients to

$$D_{13} = D_1, \ D_{23} = D_2, \ D_{12} = \frac{D_1 D_2}{D_3},$$

where from table I we read the self-diffusion coefficient of low-dilution KCl in water as $D_1 = 1.91 \cdot 10^{-5} \, cm^2/s$, and for sucrose $D_2 = 0.52 \cdot 10^{-5} \, cm^2/s$. Here D_3 is the self-diffusion coefficient of pure water, $D_3 = 2.3 \cdot 10^{-5} \, cm^2/s$.

The initial concentrations on the top and bottom are determined from the dimensionless number

$$R = \frac{\alpha_2 Z_2}{\alpha_1 Z_1} = 0.89.$$

The initial values of the concentrations were reported to us to be

$$Z_1^0 = 0.29 \text{ and } Z_2^0 = 0.1 \, mol/l,$$

which gives initial densities differing by about 0.17%,

$$
\begin{aligned}
\rho_{\text{top}} &= \rho_0 \left(1 + \alpha_1 Z_1^0\right) = \left(1.0 + 4.8 \cdot 10^{-2} \times 0.29\right) g/cm^3 = 1.0139 \, g/cm^3, \qquad (4) \\
\rho_{\text{bottom}} &= \rho_0 \left(1 + \alpha_2 Z_2^0\right) = \left(1.0 + 12.2 \cdot 10^{-2} \times 0.1\right) g/cm^3 = 1.0122 \, g/cm^3,
\end{aligned}
$$

which are not quite consistent with the the densities reported to us, $\rho_{\text{top}} = 1.0125 \, g/cm^3$ and $\rho_{\text{bottom}} = 1.0119 \, g/cm^3$, which differ by about 0.06%. In terms of the partial densities of the two components we have

$$\rho_1^0 = Z_1^0 M_1 = 0.0216 \, g/cm^3, \quad \rho_2^0 = Z_2^0 M_2 = 0.0342 \, g/cm^3,$$

which are also very close to the initial mass fractions since the total density is very close to $1\,g/cm^3$.

Lacking better experimental data and more consistent response from the experimentalists, for the final simulations reported in the paper we use four times larger concentrations; this preserves the dimensionless number R which is presumed to be the most important number. Specifically, we set the initial mass fractions to

$$w_1^0 = 0.0864, \quad w_2^0 = 0.1368,$$

which from the EOS gives

$$\rho_{\text{top}} = \left(\frac{0.0864}{2.81} + \frac{0.9136}{1.0}\right)^{-1} = 1.0596\,g/cm^3$$

$$\rho_{\text{bottom}} = \left(\frac{0.1368}{1.55} + \frac{0.8632}{1.0}\right)^{-1} = 1.0510\,g/cm^3$$

for a density difference of about 0.8%.The plots in the bottom panel of Fig. 1 are for times $40, 50, 60\,s$, so we run our simulations out to $60\,s$. We use a geometry of $0.8cm^2$ in the $x-y$ plane (gravity along y here), to nearly match the size of the snapshots in Fig. 1. The thickness in the z direction is $0.25mm$ (so *very* thin). Our computational domain is $256 \times 256 \times 8$ zones and we use a time step limited by a diffusive mass CFL of 0.75, i.e., $\Delta t = 0.75\Delta x^2/(6*D_{\max}) = 0.75*.003125^2/(6*1.91\times10^{-5}) = 6.39\times10^{-2}s$. We are using the inertial code, as the overdamped code has trouble converging at this aspect ratio. Note that the momentum diffusion time across $1cm$ length is $\tau_\nu \sim 1cm^2/2\cdot10^{-2}\,(cm^2/s) \sim 50s$. We can use periodic BCs along x, and reservoir along y (with reservoir values set equal to the initial values), and no-slip along z. They use some special experimental procedure to get a very flat interface at the beginning, not sure however how close to a jump profile it is (with bds advection we can do a jump also). They state the wavelength of the Y-shaped instability fingers they get is on the order of $1.3mm$, which is an important number to observe. To start off the instability in deterministic simulations we can make the concentrations random in the middle layer of cells like we did for the Kevin-Helmholtz instability. The alternative is to use a flat interface and allow for thermal fluctuations to set off initial perturbations.

References

[1] Jorge Carballido-Landeira, Philip MJ Trevelyan, Christophe Almarcha, and Anne De Wit. Mixed-mode instability of a miscible interface due to coupling between rayleigh-taylor and double-diffusive convective modes. *Physics of Fluids*, 25(2):024107, 2013.

[2] Xin Liu, Andreǒ301 Bardow, and Thijs JH Vlugt. Multicomponent maxwell- stefan diffusivities at infinite dilution. *Industrial & Engineering Chemistry Research*, 50(8):4776–4782, 2011.