Supplementary information - Early Freezing Dynamics of an aqueous foam

Bubble size distribution

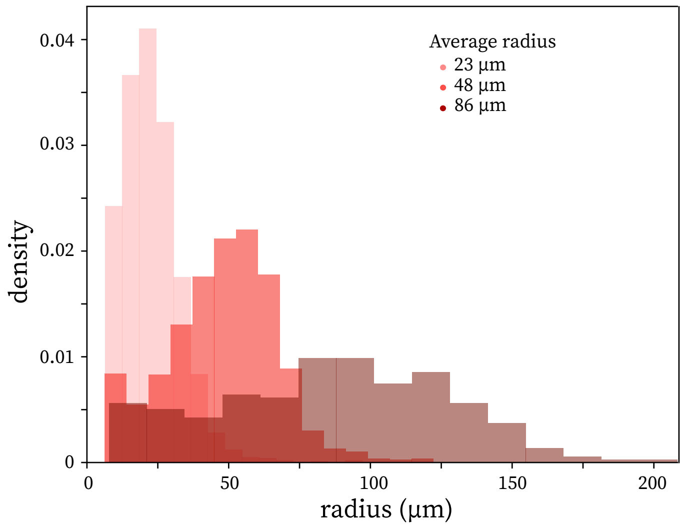

Figure 1: Distribution of the bubble radii normalized by the total bubble number for the three foams presented in Fig 2b.

Resolution of the 1D thermal problem

Given the small size of the bubbles in the studied foam compared to the observed height of the freezing front in our experiments, it is safe to assume that at a given position z in the foam column, both the gas and the liquid or solid are at the same temperature. This allows us to make an effective medium model for our thermal problem.

To predict the dynamics of the freezing front, we solve the heat equation in the three phases of our problem: solid substrate, frozen foam and liquid foam. The equations solved are :

$$(\rho C_p)_{\rm s}\frac{\partial T}{\partial {\rm t}} = \lambda_{\rm s}\frac{\partial^2 T}{\partial {\rm z}^2} \quad \text{for} \quad z \leq 0$$

$$(\rho C_p)^s_{\rm foam}\frac{\partial T}{\partial {\rm t}} = \lambda^s_{\rm foam}\frac{\partial^2 T}{\partial {\rm z}^2} \quad \text{for} \quad 0 \leq z \leq h(t)$$

$$(\rho C_p)^l_{\rm foam}\frac{\partial T}{\partial {\rm t}} = \lambda^l_{\rm foam}\frac{\partial^2 T}{\partial {\rm z}^2} \quad \text{for} \quad h(t) \leq z$$

The boundary conditions are the same as[1], meaning: $T(z \to -\infty) = T_{\rm s}$; $T(z = h(t)) = T_{\rm m}$; $T(z \to +\infty) = T_0$.

Assuming constant and uniform volume liquid/solid fraction in the liquid and solid foam, the thermal conductivities of the liquid and solid foam are given by Equation (2) in the paper. The volumetric heat capacity of the foam $(\rho C_p)^{l,s}_{\rm foam}$ is simply the volumetric heat capacity of each component of the foam weighed by its volume fraction: $(\rho C_p)^{l,s}_{\text{foam}} = \phi_l (\rho C_p)_{l,s} + (1-\phi_l)(\rho C_p)_a$ where the subscripts l, s and a stand respectively for liquid, solid and air. For simplicity of notation, we introduce the effusivity of the solid foam $e^s_{\text{foam}} = \sqrt{\lambda^s_{\text{foam}} (\rho C_p)^s_{\text{foam}}}$. The effusivities of the liquid foam and solid substrate are defined similarly. The Stefan condition controlling the propagation of the freezing front is:

$$\phi_l \rho_s \mathcal{L} \frac{\mathrm{d}h}{\mathrm{d}t} = \lambda^s_{\text{foam}} \frac{\partial T}{\partial z}(h^-) - \lambda^l_{\text{foam}} \frac{\partial T}{\partial z}(h^+)$$

This set of equations admits a self-similar solution with respect to the variable $\frac{z}{2\sqrt{D^s_{\text{foam}} t}}$ known as the Schwartz solution[1]. It requires that the position of the freezing front goes as $h(t) = 2\alpha\sqrt{D^s_{\text{foam}} t}$. The resulting temperature fields are:

$$T(z,t) = T_c + (T_c - T_{\mathrm{s}})\mathrm{erf}(\frac{z}{2\sqrt{D_{\mathrm{s}} t}}) \quad \text{for} \quad z \leq 0$$

$$T(z,t) = T_c + \frac{e_{\mathrm{s}}}{e^s_{\text{foam}}}(T_c - T_{\mathrm{s}})\mathrm{erf}(\frac{z}{2\sqrt{D^s_{\text{foam}} t}}) \quad \text{for} \quad 0 \leq z \leq h(t)$$

$$T(z,t) = \frac{T_f - T_0 \mathrm{erf}(\alpha\sqrt{\frac{D^s_{\text{foam}}}{D^l_{\text{foam}}}})}{1 - \mathrm{erf}(\alpha\sqrt{\frac{D^s_{\text{foam}}}{D^l_{\text{foam}}}})} + \frac{T_0 - T_f}{1 - \mathrm{erf}(\alpha\sqrt{\frac{D^s_{\text{foam}}}{D^l_{\text{foam}}}})}\mathrm{erf}(\frac{z}{2\sqrt{D^l_{\text{foam}} t}}) \quad \text{for} \quad h(t) \leq z$$

With the contact temperature at $z = 0$ being:

$$T_c = \frac{T_f + \frac{e_s}{e_g}\mathrm{erf}(\alpha) T_s}{1 + \frac{e_s}{e_g}\mathrm{erf}(\alpha)} \tag{1}$$

The value of α is given by the Stefan condition which becomes:

$$\alpha = S_{\mathrm{t}} \frac{e_{\mathrm{s}}}{e^s_{\text{foam}}} \frac{e^{-\alpha^2}}{\sqrt{\pi}(1 + \frac{e_{\mathrm{s}}}{e^s_{\text{foam}}}\mathrm{erf}(\alpha)} - \gamma S_{\mathrm{t}} \frac{e^l_{\text{foam}}}{e^s_{\text{foam}}} \frac{e^{-\alpha^2 D^s_{\text{foam}}/D^l_{\text{foam}}}}{\sqrt{\pi}(1 - \mathrm{erf}(\sqrt{\frac{D^s_{\text{foam}}}{D^l_{\text{foam}}}}\alpha))} \tag{2}$$

For more concise notation, we introduce $\gamma = \frac{T_0 - T_m}{T_m - T_{\mathrm{s}}}$. The Stefan number is now defined as $\mathrm{S_t} = (\rho C_p)^s_{\text{foam}}(T_m - T_{\mathrm{s}})/\phi_l \rho_s \mathcal{L}$ with $\mathcal{L}$ the latent heat of solidification of the water solution and ranges between $\mathrm{S}^{min}_{\mathrm{t}} = 0.09$ and $\mathrm{S}^{max}_{\mathrm{t}} = 0.65$ in our experiments. Equation (2) is solved numerically for α to compute $h(t)$.

We note that the contact temperature (1) between the substrate and the frozen layer has the same expression as found by Thiévenaz *et al.* but the parameter α now depends on the initial temperature T_0. Moreover, for large values of $\gamma = \frac{T_0 - T_m}{T_m - T_{\mathrm{s}}}$, the transcendental equation does not have a solution. This is because the initial temperature of the liquid phase is too hot and heats the substrate above T_{m}. In that case, freezing does not occur.

Bibliography

[1] P. Kant, R. B. Koldeweij, K. Harth, M. A. van Limbeek and D. Lohse, *Proceedings of the National Academy of Sciences*, 2020.

[2] V. Thiévenaz, T. Séon and C. Josserand, *J. Fluid Mech.*, 2019, **874**, 756–773.