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The formation of filamentary structures from molten silicates: Pele's hair, angel hair, and blown clinker

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ABSTRACT

We conduct an analysis of the concomitant, competing phenomena at play in the formation of long filamentary structures from a stream of hot, very viscous and cohesive liquid as it is blown by a fast, cool air stream. The situation is relevant to a broad class of problems, namely volcanic glass threads or fibers formed when small particles of molten material are thrown into the air and spun out by the wind into long hair-like strands (called Pele's hair), to the process of prilling, the manufacture of glass fibers, and the formation of coke in furnaces and combustion chambers.
The air stream blowing on the molten material both breaks up the liquid into fragments stabilized by capillarity, and cools the liquid down to solidification. There are, in this problem, four characteristic times. First, a deformation time of the liquid masses, setting the rate at which drops elongate into ligaments. Then, two timescales set the time of capillary breakup of these ligaments, one prevailing on the other depending on the relative weight of inertia on viscous slowing (that point is illustrated by an original experiment). Finally, a solidification time of the ligaments. Thin solid strands will only form when solidification occurs before capillary breakup. We have discovered that this condition is likely to apply when the liquid is strongly viscous, as for clinker in the cement industry, considered here as a generic example. We formulate recommendations to remove (or enhance) the formation of these objects.

1. Introduction

Pele's hair (named after Pele, the Hawaiian goddess of volcanoes) is a geological term primarily used by volcanologists for volcanic glass threads or fibers formed when small particles of molten material are thrown into the air and spun out by the wind into long hair-like strands (Fig. 1). The diameter of the strands is typically a fraction of a millimeter, and they can be as long as meters [1–4].

The phenomenon is also known in the context of *prilling*. A prill is a small aggregate of a material, most often a dry sphere, formed from a melted liquid [5]. The material to be prilled is a solid at room temperature and ideally a low viscosity liquid when melted. Prills are formed by allowing drops of the melted prill substance to congeal, freeze or evaporate a solvent in mid-air after being dripped from the top of a prilling tower, possibly through a counter current of air. Melted material may also be atomized and then allowed to form smaller prills that are useful in cosmetics, food, and animal feed. The process is known to form undesirable thin strands, named *angel hair* in this context, when the initial viscosity of the liquid is too high.

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Fig. 1. Top: Lava fountaining in Hawaiian type volcanoes: Exploding lava bubbles (tephra) are torn into thin glassy threads, so-called Pele's hair (Nyiragongo volcano, Congo, January 2011). Middle: Pele's hair refers to extremely long sub-millimetric threads of brownish colored basalt glass (sideromelane and tachylite). Note that a ligament was in the process of breaking up into drops as it has solidified. Bottom: Millimetric drops, named achneliths (also known as Pele's tears) are often found attached to one end of individual threads. The longest dimension of specimen is 9 mm.

Clinker, a mixture of limestone (calcium carbonate $CaCO_3$) and clay (silica SiO_2 with an admixture of alumina Al_2O_3), is the major component if cement. The mixture is melted in an oven at high temperature (1450 °C) into a liquid of high viscosity, and high surface tension. Because it is more easily transported in its solid phase when it is conditioned in small (millimetric) pellets than in heavy ingots, it has been found convenient to atomize the stream of liquid clinker at the exit of the oven by a fast cold air jet. This process has two purposes, first to atomize the liquid at the desired size, then to cool the stable drops down to the freezing temperature, in order to obtain solid pellets, which are further collected. However, it has also been noticed that in this course of events, long thin threads were occasionally formed. These are, for obvious sanitary reasons, undesirable.

In other applications such as the preparation of *cotton candy* (*candyfloss*), or the manufacture of glass fibers, they are on the contrary the desired objects, whose features need to be controlled.

Aiming at understanding why and how these long filamentary objects occur and are formed, we give below a scaling analysis of the concomitant breakup and solidification phenomena at play, leading to orders of magnitude estimates for the lengths and time scales involved in this problem. We also provide recommendations for removing them in the particular case of blown clinker, which we take as a generic example.

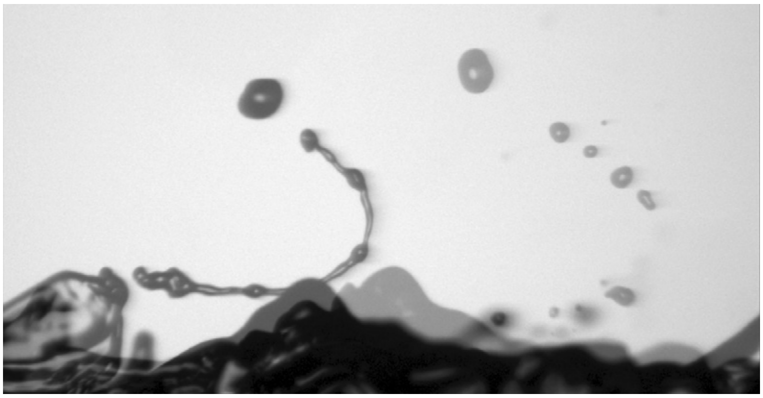

Fig. 2. A centimetric in length water ligament stretched in a fast ($\simeq 70$ m/s) air stream blowing parallel to the liquid bulk surface, breaking up as it detaches from the bulk.

2. Molten silicates: highly viscous and cohesive liquids

As it exits the oven of a cement factory, clinker is a hot liquid, with unusual properties: it is extremely viscous (a thousand times more than water), and cohesive (like mercury), properties that this material shares with molten soda-lime glasses, and volcanic tephra [4].

- Initial temperature: $\theta_0 = 1450\,^\circ\mathrm{C}$;
- Solidification temperature: $\theta_s = 700\,^\circ\mathrm{C}$;
- Viscosity at θ_0: $\eta = 2500 \times 10^{-3}$ Pa s (two thousand times that of water);
- Surface tension: $\sigma = 0.5$ N/m (nearly ten times that of water, like mercury);
- Density: $\rho = 2300$ kg/m^3;
- Thermal diffusivity: $\kappa = \lambda/\rho C_p = 3 \times 10^{-6}$ m^2/s.

It is atomized by a fast cold air jet whose transverse size is much larger than that of the clinker stream (of the order of ten centimeters), trickling down from a pipe in a cross flow, and which therefore atomizes and disperses in a nearly infinitely diluted fashion. The useful air stream features are:

- Velocity: $u \simeq 100$ m/s;
- Temperature: $40\,^\circ\mathrm{C}$;
- Density: $\rho_a \simeq 1$ kg/m^3;
- Kinematic viscosity: $\nu_a \simeq 10^{-5}$ m^2/s;
- Thermal diffusivity: $\kappa_a \simeq 10^{-5}$ m^2/s ($Pr = \mathcal{O}(1)$).

3. Initial fragmentation and kinematics

The liquid clinker effluent is, in most practical conditions, first violently atomized by an air stream, which then will also cool it. The same phenomenon occurs with lava, explosively atomized at the exit of a volcano (see Fig. 1). Indeed, the large air inertia $\rho_a u^2$ overcomes easily the liquid cohesion, via a series of instabilities (see [3] for a review on the destabilization of a liquid jet in a cross flow) ending when the liquid lumps have a sufficiently small size d_0 so that capillary confinement σ/d_0 (owing to Laplace law) equilibrates inertial destabilization, namely [6]

$$d_0 \sim \frac{\sigma}{\rho_\mathrm{a} u^2} \tag{1}$$

thus setting the *largest* admissible liquid drop diameter after the primary instabilities have chopped off the liquid effluent (see [7] for a detailed discussion in a related context, including the resulting fragment size distribution). We concentrate in situations dominated by a mean permanent velocity difference u seen by all the particles, irrespective of their size, and do not consider scale-dependent velocity spectra, which would suit more to turbulent fields (see [8–11]). As long as it remains attached to the liquid bulk, a spheroidal liquid lump further stretches in the flow in the form of a ligament (as seen in Fig. 2) with length $\ell(t)$ and diameter $d(t)$ such that its initial volume d_0^3 is conserved,

$$\ell(t)\, d(t)^2 \sim d_0^3 \tag{2}$$

at a rate given, at short times by (see [12–14,7])

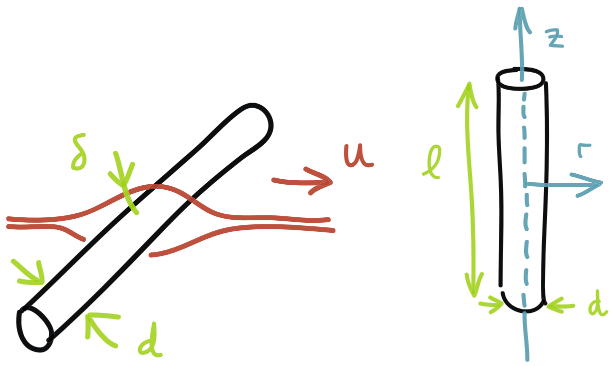

Fig. 3. Sketch of the boundary mayer development past a ligament of size d, and of system of coordinates $\{r, z\}$. The ligament is stretched in the z direction.

$$\ell(t) = d_0(1+\gamma t)^2 \quad \text{with } \gamma = \frac{u}{d_0}\sqrt{\frac{\rho_a}{\rho}} \tag{3}$$

so that

$$d(t) = d_0(1+\gamma t)^{-1} \tag{4}$$

The elongation rate γ is based on the destabilizing stream velocity u minored by the factor $\sqrt{\rho_a/\rho}$ reflecting the efficiency of the transfer of momentum from the gas to the liquid phase. Since $\rho_a/\rho \ll 1$, the initial stretching velocity of the liquid ligaments is much smaller than u. Viscous stresses in the liquid are subdominant in the initial deformation phase.

4. The thermal problem

The liquid is initially very hot, and therefore cools in the air blowing on it. The blowing velocity is given by the contrast between the stretching velocity $u\sqrt{\rho_a/\rho}$ and u itself, and it therefore of order u. A boundary layer thus forms around the stretching ligament (see Fig. 3), with thickness

$$\delta = \sqrt{\frac{\nu_a d_0}{u}} \tag{5}$$

For the ligament to cool, heat must be transported radially through the liquid, and then cross the boundary at its surface. Which step is the limiting one? The answer to this question amounts to consider the relative magnitude of the heat conduction times in the liquid bulk d_0^2/κ, and in the thermal *sheared* boundary δ_θ^2/κ_a (where $\delta_\theta = \delta\, Pr^{-1/3} \approx \delta$, see e.g. [15] since $Pr = \nu_a/\kappa_a = \mathcal{O}(1)$ in gases). Obviously conduction in the liquid is the slowest process,

$$\frac{d_0^2}{\kappa} \gg \frac{\delta^2}{\kappa_a} \tag{6}$$

since

$$\frac{ud_0}{\nu_a} \gg \frac{\kappa}{\kappa_a} = \mathcal{O}(10^{-1}) \tag{7}$$

The removal of heat at the ligament surface is indeed extremely efficient. However, diffusion takes place on a moving substrate in the ligament, which is continuously stretched, and thus compressed radially as it cools. Therefore, the real problem to analyze is a *stretching enhanced* diffusion process in the ligament. Indeed, the characteristic time of the ligament's diameter shrinking γ^{-1} is shorter also than d_0^2/κ, the ratio of which forming a Péclet number

$$Pe = \frac{\gamma d_0^2}{\kappa} \tag{8}$$

which is much larger than unity in the present case. On a moving substrate, the heat conservation equation is

$$\partial_t\theta + \boldsymbol{u}\cdot\nabla\theta = \kappa\nabla^2\theta \tag{9}$$

and simplifies if written in a moving frame $\{r, z\}$ whose axes are aligned with the directions of maximal compression and stretching of the elongating ligament where $\boldsymbol{u} = \{(\dot{d}/d)r, -2(\dot{d}/d)z\}$, according to

$$\partial_t\theta + (\dot{d}/d)r\partial_r\theta = \kappa\,\partial_r(r\partial_r\theta)/r \tag{10}$$

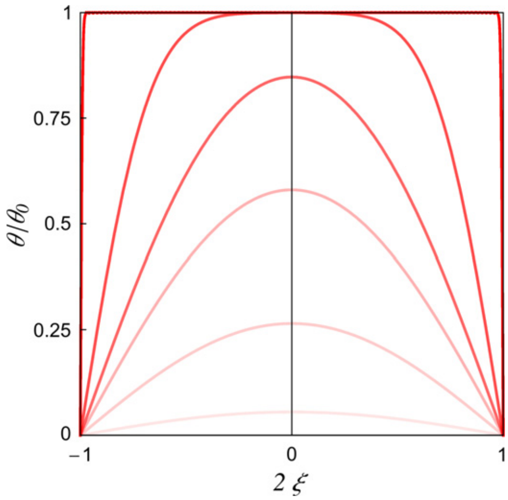

Fig. 4. Evolution of the temperature field within a compressed slab for successive times $\tau = 10^{-4}$ (the initial top hat profile), 0.1, 0.5, 1, 2, 4.

where the (vanishingly small) longitudinal temperature gradient $\partial_z\theta$ has been discarded, expressing that temperature gradients are essentially zero along the stretched direction z of the ligament, and concentrate in the radial direction r. This equation, where stretching appears through the apparent elongation rate $(\dot{d}/d)$ only, is solved in closed form (see [16], and references therein), by making use of the following transformations

$$\xi = \frac{r}{d(t)}, \quad \text{and} \quad \tau = \kappa \int_0^t \frac{\mathrm{d}t'}{d(t')^2} \tag{11}$$

leading to a pure diffusion equation in these units of time and space

$$\partial_\tau\theta = (\partial_\xi\theta)/\xi + \partial_\xi^2\theta \tag{12}$$

Willing to avoid Bessel functions, we consider for technical simplicity the two dimensional version of the problem, strictly valid for a slab, the underlying physics being obviously identical

$$\partial_\tau\theta = \partial_\xi^2\theta \tag{13}$$

whose solution, starting from a uniformly hot ligament at temperature θ_0 relative to its cooling environment is [17]

$$\frac{\theta}{\theta_0} = \frac{2}{\pi}\sum_{n=0}^{\infty}\frac{(-1)^n}{n+\frac{1}{2}}\cos\left[\left(n+\frac{1}{2}\right)\pi\xi\right]e^{-(n+\frac{1}{2})^2\pi\tau} \tag{14}$$

with

$$\tau \sim \frac{(\gamma t)^3}{Pe} \quad \text{with } Pe \gg 1 \tag{15}$$

The temperature profiles for different successive times are displayed in Fig. 4. The maximal temperature in the ligament in $r = 0$ decays as

$$\theta(\xi = 0, \tau) \simeq \theta_0\frac{4}{\pi}e^{-\pi\tau/4} \tag{16}$$

and the ligament has solidified when this temperature has decayed down to the solidification temperature θ_s. More precisely, the viscosity of the melt increases sharply as its temperature decays toward θ_s (see for instance [18]). We simply consider that the ligament is a solid when its core temperature is θ_s, and was a standard viscous liquid before, giving

$$\tau_s \sim \ln(\theta_0/\theta_s) \tag{17}$$

and thus providing finally the solidification time t_s as

$$\gamma t_s \sim \big(Pe\ln(\theta_0/\theta_s)\big)^{1/3} \tag{18}$$

This time is essentially fixed in order of magnitude by the time it takes for the ligament to deform γ^{-1}, the intrinsic diffusive properties of the liquid being embedded in the Péclet number through a weak (1/3) power law correction.

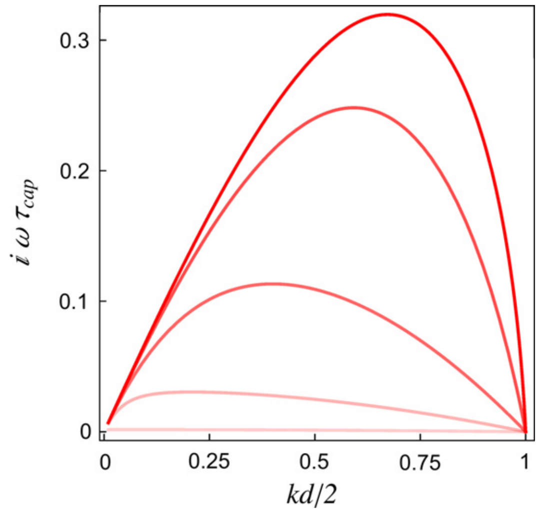

Fig. 5. Dispersion curve for $Oh = 0.05$ (thick line), 0.2, 1, 5, 100 (thin line).

5. Ligament breakup, or not

5.1. Basics

Liquid thread breakup is a capillary driven instability which occurs thanks to the near cylindrical shape of the ligaments (see [3] for an extensive review). When liquid surface tension and inertia are solely at play, the ligament-drops topology change is completed within a characteristic time given by

$$t_{\text{cap}} \sim \sqrt{\frac{\rho d_0^3}{\sigma}} \tag{19}$$

while if viscous forces dominate over inertia, they slow down the process (without suppressing it) which now occurs on a typical timescale

$$t_{\text{vis}} \sim \frac{\eta d_0}{\sigma} \tag{20}$$

The ratio of these two timescales defines the Ohnesorge number as

$$\frac{t_{\text{vis}}}{t_{\text{cap}}} = Oh \quad \text{with } Oh = \frac{\eta}{\sqrt{\rho d_0 \sigma}} \tag{21}$$

A convenient form of the dispersion relation (when perturbations of the ligament diameter are expanded proportionally to $\exp(ikz - i\omega t)$) describing the range of unstable wavelengths and associated growth rate (for a nonstretched ligament) is

$$(-i\omega)t_{\text{cap}} = \sqrt{\frac{1}{2}\left(x^2 - x^4\right) + \frac{9}{4}Oh^2 x^4} - \frac{3}{2}Oh\, x^2 \quad \text{with } x = kd_0/2 \tag{22}$$

displaying the most amplified wavenumber

$$k_m d_0/2 = \frac{1}{\sqrt{2 + 3\sqrt{2}Oh}} \tag{23}$$

going to zero like $1/\sqrt{Oh}$ (and thus producing more distant drops along the ligament as viscosity increases), with associated growth rate

$$-i\omega(k_m)t_{\text{cap}} = \frac{1}{2\sqrt{2} + 6Oh} \tag{24}$$

Fig. 5 shows the deformation of the dispersion curve as the Ohnesorge number is varied. In the absence of stretch, the ligament breakup characteristic time thus switches from t_{cap} to t_{vis} as the liquid viscosity increases.

With longitudinal stretch, a similar analysis (see [3]) shows that as long as the deformation time of the ligament γ^{-1} is shorter than the relevant instability time t_{cap} or t_{vis}, instability is suppressed (conversely, if γ^{-1} is larger than the instability time, destabilization of a continuously stretched ligament occurs within γ^{-1}). The phenomenon is clearly apparent in Figs. 6

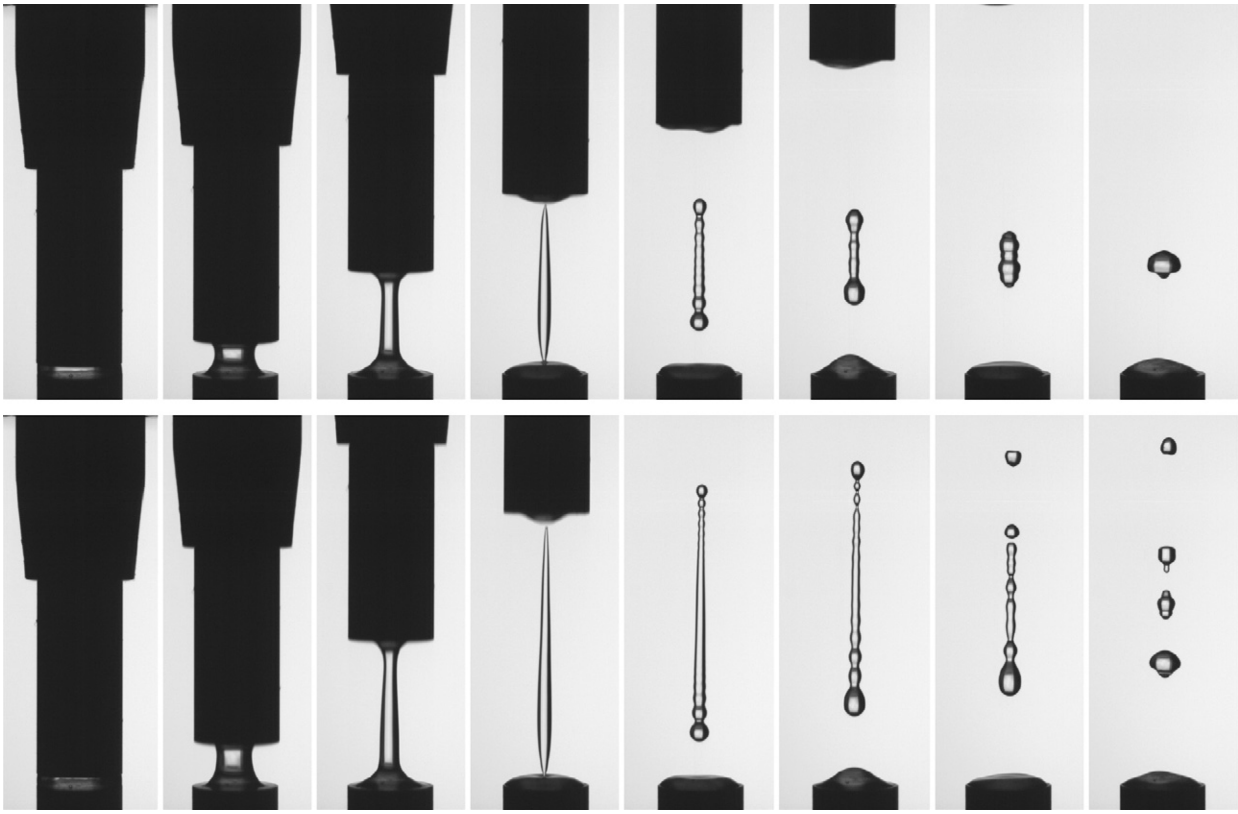

Fig. 6. A volume of water is initially squeezed between two facing solid rods of diameter $d_0 = 8$ mm ($Oh = 1.3 \times 10^{-3}$, $t_{\mathrm{cap}} = 0.1$ s), which are then separated with an elongation rate γ such that $\gamma t_{\mathrm{cap}} = \sqrt{We} > 1$. The pictures are spaced by 25/3000 s. Top: $\gamma = 100\ \mathrm{s}^{-1}$. Bottom: $\gamma = 175\ \mathrm{s}^{-1}$. Note that the ligament separates from the rods at the same instant of time (fourth image) in both cases, independent of γ.

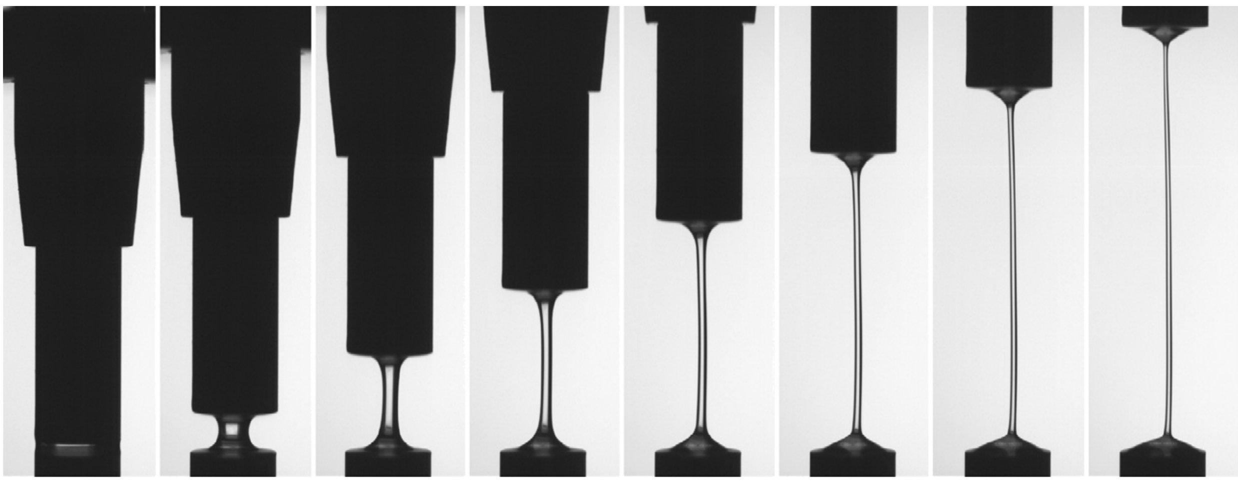

Fig. 7. Same as in Fig. 6 except that the liquid is now a silicone oil 100 times more viscous than water (and comparable surface tension). The corresponding breakup time $t_{\mathrm{vis}} = 1$ s is so long that the ligament goes on stretching and never detaches from the rods. If it were hot and liable to solidify, it would have time to cool and form a solid strand before breaking up: the Pele's hair.

and 7: a stretched liquid ligament remains smooth as long as its extremities are attached to the separating solid rods pulling them. However, as soon as the extremities of the ligament have detached from the pulling rods, it is therefore no more stretched, and destabilizes in a timescale given by its current diameter (hence all the more rapidly that it has been strongly elongated before).

Remarkably, the time it takes for the extremities to detach is independent of the rate at which the ligament is stretched γ, and is solely set by t_{cap} (or t_{vis}) as seen in Fig. 6 (or Fig. 7). Indeed, the stretch vanishes at the extremities (presumably on an axial distance of the order of the rods diameter), which are pinned at the rods surface. These regions at the extremities thus evolve on their own, basically insensitive to the rest of the (stretched) ligament.

5.2. Discussion

We have now all the required ingredients to discuss the conditions for the formation of long thin stands: ligaments will be stretched for a period lasting t_{cap}, or t_{vis} depending on Oh. We will thus have to compare these timescales to the solidification timescale t_s to see if, when and how solid elongated filaments are formed.

In the case of blown clinker, we have

$$\gamma t_{\mathrm{cap}} \sim \sqrt{We} \quad \text{with } We = \frac{\rho_a u^2 d_0}{\sigma} = \mathcal{O}(1) \tag{25}$$

by definition of the initial maximal admissible fragment diameter $d_0 \approx 5$ mm. Let us start to construct the ratio of t_s to t_{cap}, we have (taking $\tau_s = \mathcal{O}(1)$)

$$\frac{t_s}{t_{\mathrm{cap}}} \simeq \frac{Pe^{1/3}}{\sqrt{We}} \sim Pe^{1/3} \tag{26}$$

which is, owing to the value of the Péclet number in the present case

$$Pe = \frac{\sigma}{\sqrt{\rho\rho_a} u\kappa} = \mathcal{O}\left(10^2\right) \tag{27}$$

larger than unity. Indeed,

$$\frac{t_s}{t_{\mathrm{cap}}} \approx 4 \tag{28}$$

In other words, if the relevant timescale of the capillary instability is t_{cap}, then the stretching ligaments will take too long to cool, and will breakup into stable drops before they solidify. One expects in that case the formation of solid pellets, the Pele's tears, but there will be no Pele's hair attached to them in that limit.

Conversely, if the liquid is initially sufficiently viscous so that t_{vis} now sets the instability timescale, which is believable when

$$Oh = \frac{\eta u}{\sigma}\sqrt{\frac{\rho_a}{\rho}} \tag{29}$$

is larger than unity, as in the case of clinker for which $Oh = 10$, then

$$\frac{t_s}{t_{\mathrm{vis}}} \simeq \frac{Pe^{1/3}}{Oh} \approx 0.4 \tag{30}$$

The conclusion is now opposite. Ligaments take so long to breakup that they have time to cool, and solidify. They do so in thin strands of diameter

$$\frac{d_s}{d_0} \sim (1+\gamma t_s)^{-1} \approx 0.2 \tag{31}$$

roughly five times smaller than the Pele's tears to which they are attached, consistently, at least qualitatively, with what can be seen in Fig. 1.

6. Conclusions

We have conducted an analysis of the concomitant, competing phenomena at play in the formation of long filamentary structures from a stream of hot, very viscous and cohesive liquid as it is blown by a fast, cool air stream, and have examined the case of molten clinker as a generic example. The air stream both breaks up the liquid into fragments stabilized by capillarity, and cools the liquid down to solidification.

There are, in this problem, four characteristic times. First, γ^{-1} which sets the deformation time of the liquid masses in the mixture, and the rate at which drops elongate into ligaments. Then, two timescales (t_{cap} and t_{vis}) set the time of capillary breakup of these ligaments, depending on the relative weight of inertia on viscous slowing. Finally, t_s is the cooling time of the ligaments.

Thin solid strands will only form when t_s is shorter than the relevant capillary instability time. We have discovered that this condition is likely to apply when the liquid is strongly viscous, as for clinker. Some of the fragments of Volkhovites [19,20] in Fig. 8 are presumably an interesting marginal case for which t_s is of order t_{vis} since they have the characteristic dumbbell shape of drops which have solidified as they were in the process of detaching.

When they are not desired, namely in the cement industry for example, the only way to remove these long thin objects is to increase the ratio t_s/t_{vis}, and make it larger than unity. To this end, there are several options:

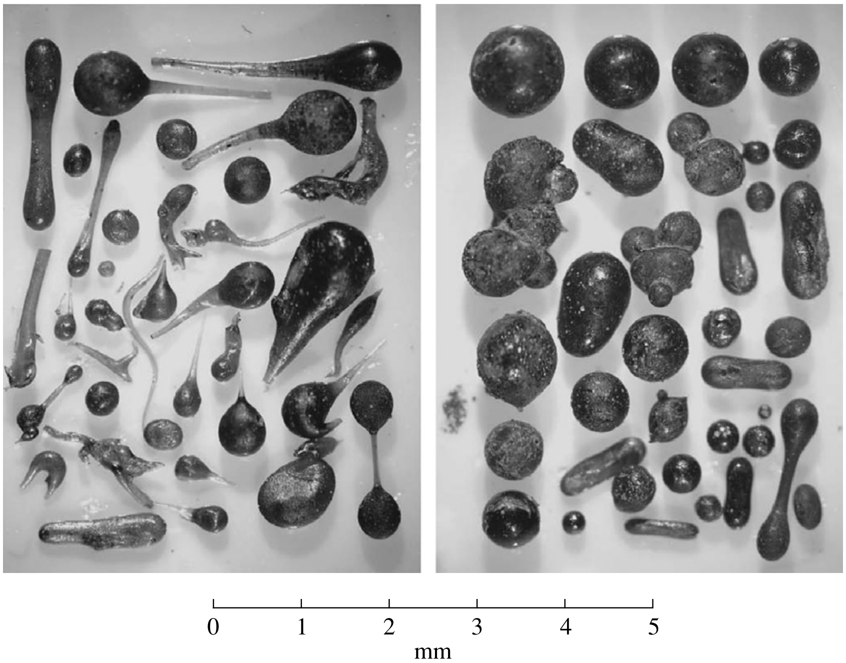

Fig. 8. Volkhovite (a kind of tektites, 10 000 years old, found near the river Volkhov in Russia) grains of various shapes and colors. Note the dumbbell shape indicating that most probably, t_s/t_{vis} was of order unity when these objects formed. Adapted from [19].

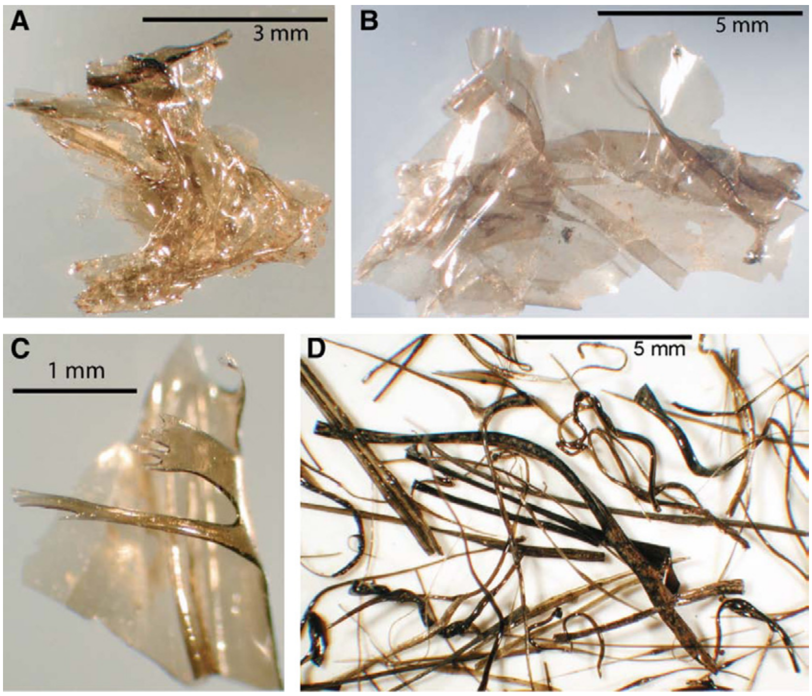

Fig. 9. Products of a submarine explosion: A) and B) Intensely folded limu o Pele fragments only 5 µm thick; C) Thin curved limu o Pele fragment that stretched beyond the tensile strength of the melt, which pulled apart; D) Selection of Pele's hair fragments from the summit of Axial Seamount on the Juan de Fuca Ridge, many are curved and some are tack-welded to themselves. Adapted from [21].

(i) Increase t_s by blowing with hot air, but this would require to use an air temperature nearly equal to that of clinker, namely 1000 °C or so, which does not look reasonable.
(ii) Decrease the Ohnesorge number by starting with a less viscous liquid. This option is standardly adopted in the *prilling* context: diluting the solution to be atomized with more solvent removes the *angel hairs*. But this looks difficult in practice since clinker is as it is and there is no simple way to dilute it.

(iii) In the same vein, the Ohnesorge number can be decreased by using a slower air stream velocity u. This will also have the effect of increasing the initial maximal size d_0 (which depends strongly on u), and therefore to lead to thicker stands if they form. Decreasing u from 100 m/s to 50 m/s should already have an effect.

To close, let us mention that these long strands are also transitorily formed in hydromagmatic (see [22] and references therein) undersea volcanic eruptions (consistent with our analysis pointing out the importance of the factor $\sqrt{\rho_a/\rho}$ in the Ohnesorge number, much larger when the destabilizing phase is a liquid). The solidification of the lava bubble can be so fast that it solidifies as thin *sheets* (called limu o Pele, see Fig. 9, and top Fig. 1 for comparison). However, they are typically not collected in the resulting fragments, which are coarse brittle shattered after solidification by the repeated wave action when they are thrown back to the beach. Pele's hair are indeed fragile objects which do not resist further crushing and grinding. This maybe also suggests a way to sweep them out in the industry context.

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