UNJAMMING STRONGLY COMPRESSED PARTICLE RAFTS

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ABSTRACT

We experimentally study the unjamming dynamics of strongly compressed particle rafts confined between two fixed walls and two movable barriers. The back barrier is made of an elastic band, whose deflection indicates the local stress. The front barrier is pierced by a gate, whose opening triggers local unjamming. The rafts are compressed by moving only one of the two barriers in the vicinity of which folds form. Using high speed imaging, we follow the folded, jammed, and unjammed raft areas and measure the velocity fields inside and outside of the initially confined domain. Two very different behaviors develop. For rafts compressed by the back barrier, only partial unjamming occurs. At the end of the process, many folds remain and the back stress does not relax. The flow develops only along the compression axis and the particles passing the gate form a dense raft whose width is the gate width. For rafts compressed at the front, quasi-total unjamming is observed. No folds persist and only minimal stress remains, if any. The particles flow along the compression axis but also normally to it and form, after the gate, a rather circular and not dense assembly. We attribute this difference to the opposite orientation of the force chain network that builds up from the compressed side and branches. For rafts compressed at the gate side, keystone particles are immediately removed which enhances local disentanglement and leads to large scale unjamming. In contrast, for back compressed rafts, the force chain network redirects the stress laterally forming arches around the gate and resulting in a limited unjamming process.

1 Introduction

Capillary adsorbed particles at liquid interfaces have been since long used in emulsions and foams [Ramsden (1903); Pickering (1907); Pitois and Rouyer (2019)]. Their potential for further applications, especially for producing bijels [Binks (2002a); Herzig et al. (2007); Cates and Clegg (2008)], chemical-free reversible encapsulation [Aussillous and Quéré (2001); Abkarian et al. (2013); Jambon-Puillet et al. (2018); Pike et al. (2002)] and membranes [Krachevsky et al. (2001)] has renewed interest in them. In this context, one key-attribute of the particles is their capacity to stabilize interfaces. This stabilization partly results from the large reduction of surface energy caused by the particle adsorption. Further stabilization originates from the physical barrier the particles build that prevents direct contact with other surfaces. The effects resulting from these two stabilization mechanisms remain challenging to characterize at the macroscopic scale. The most common way consists in characterizing these effects via the description of the interfacial mechanical properties. Thanks to intensive research, these properties are fairly well outlined for quasi-static regimes and moderate compression. Yet, for general conditions, they remain poorly understood and therefore poorly predictable. This lack of knowledge is particularly profound for large strains and large strain rates or when strong gradients of particle density exists. For these conditions, commonly encountered in natural situations and industrial processes [Garbin (2019)], barely no data exist. Consequently, key aspects such as self-healing capacity, self-healing dynamic and processability of these interfaces remain widely unexplored. Our paper aims to shed some light onto these topics.
Before detailing our method and findings, it is helpful to recall the existing knowledge about the mechanical properties of particle-laden interfaces. At moderate particle density, the interface is viscous. It becomes viscoelastic upon the particle network percolation, and finally behaves as a solid for larger particle density [Reynaert et al. (2007); Cicuta et al. (2003); Lagubeau (2010)]. The transition toward a solid-like behavior is associated to a pressure collapse [Aveyard et al. (2000); Monteux et al. (2007)] and attributed to a jamming process [Liu and Nagel (1998)], a phenomenon common to other athermal assemblies, also called soft glassy materials [Sollich et al. (1997); Hebraud and Lequeux (1998)]. If further compressed, the interface buckles giving rise to regular wrinkles whose amplitude regularly increases, until one of them dramatically grows and eventually collapses into a large fold.

To date, most studies were motivated by practical interest such as bubble dissolution arrest and foam stabilization [Binks (2002b); Garbin (2013)]. While this aspect is positive to arrest bubble dissolution, it remains challenging for expending interfaces. A good strategy to efficiently overcome this challenge might be to establish particle reservoirs outside the bulk, and more particularly in interfacial folds. In this context, the questions can be reformulated as such: can particles located within the sides. The paper ends with the conclusions which underline the consequences of our findings in terms of interface processability, self-healing capacity, and dynamic.

2 Experimental methods

2.1 Set-up

As sketched in Fig. [1] the experimental set-up consists of a rectangular trough with two fixed parallel walls separated by 6 cm and two movable barriers that can be translated along the two walls to compress the confined particle raft. The
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Figure 1: Experimental set-up sketched from (a) top and (b) side view. The particle raft relaxed length, $L_r$, is equal to its width $W$. The raft is compressed by moving either the back barrier (red) or the front one (blue) to obtain a length $L_c < L_r$. The elastic deflection $\delta$ (exaggerated here) enables to calculate the local stress.

so-called "back" barrier is made of an elastic string placed in the plane of the interface, perpendicularly to the side walls. The string is produced in house by injecting a (1:1) mixture of Elite Double 8 basis and catalyst (Zhermack Spa) into a glass capillary, which is manually removed after the elastomer reticulation has been completed. The elastic is then fixed to a 6 cm broad structure and calibrated using known weights to obtain its Young modulus, for details please see Appendix. During the experiments, the elastic deflection, $\delta$, is measured and used to calculate the stress developing at the back side of the raft, see Appendix. The second barrier, refereed to as "front" barrier, is pierced in its center by an orifice of width $w = 1 cm$. A vertically sliding gate made of a thin plate enables to maintain the orifice closed. By suddenly lifting the gate, the orifice gets unblocked and the applied stress is locally released.

2.2 Particles

The particles are sieved and silanized glass beads. The diameter distribution measured over pictures of more than 1000 particles corresponds to a Gaussian with a mean of 107 $\mu m$ and a standard deviation of 8.4%. After having cleaned the particles with a piranha mixture, silanization is performed using solution of trichloro-perfluoroctylsilane in anhydrous hexane. All chemicals were purchased from Sigma-Aldrich and used as received. The resulting contact angle measured on single particles placed at the apex of a pendant drop is found to be $107^{\circ} \pm 10^{\circ}$.

2.3 Experimental procedure

The first step of the experimental procedure consists in obtaining square rafts. To do so, the trough is filled with distilled water and particles are sprinkled on the interface between the barriers. By gently blowing on them, we insure that they distribute in a monolayer. This monolayer is then compressed and decompressed by moving any of the two barriers and the quantity of particles is adjusted in order to have a square relaxed raft. We define the raft as relaxed as soon as the stress measured during decompression vanishes. Noting $L_r$, the relaxed length and $W$ the trough width, we have $L_r = W = 6 cm$. Once a square raft is formed, the distance between the barriers is increased and the raft is annealed by stirring the particle assembly. The raft is then compressed again, but by moving only one of the two barriers, the other one remaining fixed until the end of the experiment. The state of compression is then given by $K = (L_r - L_c)/L_r$ with $L_c$ the distance between the two barriers in their final position. Finally, the relaxation is triggered by suddenly opening the gate. A high speed camera placed under the raft records its evolution. Practically,
3000 frame per second are recorded. The images have a resolution of 14.1 µm/pixel. The movies are then analysed to provide different measurements.

![Image sequences of strongly compressed rafts](image)

Figure 2: Image sequences of strongly compressed rafts ($K = 67\%$) after local release of the stress achieved by lifting the gate of the front barrier. The opening instant is taken as time origin. The red color indicates unjammed particles area while the blue shows the “escaped” particles. Top: back compression (subscript b) with from left to right: $t_{b,1} = 70$ ms, $t_{b,2} = 120$ ms, $t_{b,3} = 250$ ms, $t_{b,4} = 360$ ms and $t_{b,5} = 430$ ms. Bottom: front compression (subscript f) with from left to right: $t_{f,1} = 80$ ms, $t_{f,2} = 410$ ms, $t_{f,3} = 630$ ms, $t_{f,4} = 1600$ ms, $t_{f,5} = 2800$ ms.

3 Measurements

3.1 Unjammed, jammed, folded and escaped areas

During the raft relaxation, several quantities are measured, which first need to be defined. The illustrative image sequences displayed in Fig. 2 are useful to do so. Let us first consider what happens in the initially confined domain. After the gate opens, some particles locally unjam. The corresponding surface area, $A_{uj}^*$, colored in red in Fig. 2, is tracked using ImageJ and the machine learning plug-in called Trainable Weka Segmentation Arganda-Carreras et al. (2017). After normalisation by the initial confined domain area $A_c = W L_c$, we obtain $A_{uj} = A_{uj}^*/A_c$. Per definition, the particles that remain jammed occupy the normalized area $A_j = 1 - A_{uj}$. They can take the form of macroscopic folds or remain in the plane of the interface. To characterize this distribution, we measure the so-called folded surface area $A_f^*$. Practically, the folds correspond to interface portions which make a significant angle to the horizontal plane and therefore appear dark on the back-lighted pictures. Thus, $A_f^*$ is obtained by using a threshold function and is then normalized by $A_c$ to provide $A_f$. Note that this term refers to the projection of the folded surface and not to the surface area contained in these folds, which is not accessible with our images.

Let us now look to what happens outside of the initially confined domain. Some of the unjammed particles flow through the orifice and migrate further to form a more or less dense assembly, colored in blue in Fig. 2. The surface area occupied by this assembly $A_e^*$, is tracked using a threshold function applied after subtraction of the background taken in the absence of particles. The normalized escaped area $A_e$ is obtained by dividing $A_e^*$ with $(L_r - L_c)W = A_r - A_c$, i.e. with the difference between the relaxed raft area and the one of the confined domain. This difference represents the surface that the excessive particles would occupy if forming a dense relaxed raft.

3.2 Back stress

Additionally, the deflection of the elastic barrier $\delta$, is recorded and used to compute $\Pi$, the lineic pressure (or stress) developing at the back of the raft. For details about the conversion of $\delta$ into $\Pi$, please read the Appendix.

3.3 Velocity fields

Finally, we use the PIVlab routine of Matlab Thielicke and Stamhuis (2014); Thielicke and Sonntag (2021) to gain information about the velocity fields developing in these systems. In practise, we perform three types of Particle Image Velocimetry (PIV).
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The first one focuses on the flow inside the confined area, and more precisely within the unjammed area. To avoid wrong interpolations with movements taking place in the jammed raft, such as fold translation, masks corresponding to the identified unjammed areas (red regions in Fig. 2) are applied, limiting the analysis to the region of interest. The second analysis is similar and consists in performing PIV outside of the confined domain using masks that correspond to the escaped assembly (blue regions in Fig. 2). Note that we further reduce the masks of about 5 mm on the orifice side, since the nonuniform background of this zone causes errors in the PIV.

These two PIVs, typically performed with a time resolution of 300 Hz, provide, for every instant - the velocity field inside and outside the confined domain. Due to the large number of frames, the instantaneous velocity fields are difficult to visualize and interpret. To facilitate their analysis, we divide each relaxation in four sub-phases, and compute the distribution of the velocity components parallel and normal to the compression axis ($v_∥$ and $v_⊥$) for each sub-phase. The sub-phases correspond to the time lapse between two consecutive images of Fig. 2, i.e., to $t_{b,i} \leq T_{b,i} \leq t_{b,i+1}$ and $t_{f,i} \leq T_{f,i} \leq t_{f,i+1}$, with $1 \leq i \leq 4$, for the back and front compression, respectively.

The third analysis deals with the velocity profile at the orifice. To obtain this information, we limit the analysis to a small zone covering the orifice and increase the time resolution to at least 600 Hz. The width of the analysed area is the orifice width and its length is fixed to 10 mm. For each picture, the obtained velocity field is then projected along the compression direction to provide the velocity profile $u(x)$, with $-w/2 \leq x \leq w/2$. The flow-rate equivalent mean velocity $\overline{u}$ is then calculated as $\overline{u} = \frac{1}{w} \int_{-w/2}^{w/2} u(x) dx$.

4 Results and discussion

Here and in the rest of this article, results obtained with front compression are presented as solid lines and full symbols while those corresponding to back compression are represented by dashed lines and empty symbols. The red color systematically indicates quantities related to the confined domain while blue is used for quantities defined outside.

4.1 Detailed analysis of rafts compressed at 67%

In this section, we present the results obtained for two rafts compressed at 67% by moving solely the front barrier or solely the back barrier. Illustrative image sequences are reproduced in Fig. 2. To facilitate the observation, the unjammed area found in the confined domain, $A_{*uj}$, is systematically colored in red and the surface occupied by the "escaped" particle assembly, $A_{*e}$, in blue. These colored masks, automatically obtained by the image treatment (section 3.3.1), are in very good agreement with the areas detected by eyes and can therefore be used for further analysis. The two rafts behave differently. For the back compression (top sequence), only a small zone found immediately behind the orifice unjams. The unjammed particles flow through the orifice, along the compression axis, to form a dense assembly whose width is the orifice width. The relaxation process stops while many folds are still visible. In contrast, the relaxation of the raft compressed from the front (bottom sequence) leads to important unjamming, which extends to almost all the confined domain. The escaped particles form a large assembly of reduced density and broad width. No folds remain indicating a full relaxation of the raft. As the caption timestamps indicate, the relaxation of the back compressed raft is much shorter. It almost stops after 0.5 s, i.e., long before the one of the front compressed raft finishes, after more than 3 s.

Figure 3: Temporal evolution of $A_{uj}$ (red), $A_f$ (black) and $A_e$ (blue) for (a) back- and (b) front- compressed raft.
To quantify this observation, we plot the temporal evolution of the normalized unjammed area $A_{uj}$ (red), folded area $A_f$ (black) and escaped assembly area $A_e$ (blue). The curves, shown in Fig. 3, confirm the qualitative findings. For the raft compressed from the back, slightly less than 15% of the initially confined area unjams and approximately 35% of the confined area appears folded at the end of the process. Interestingly, the unjammed area does not significantly grow but rather fluctuates around its mean value. These fluctuations originate the folds dynamics. The folds, initially found at the back ($t_{b,1} = 70\, ms$) migrate towards the orifice and transiently reduce the unjammed area ($t_{b,2} = 120\, ms$) before disintegrating ($t_{b,3} = 250\, ms$), explaining likewise the correlation between the local minimums of $A_{uj}$, at $\approx 0.2s$, and the local maximums of $A_f$. Finally, the particles that escape covers between 15% and 20% of the surface excessive particles or particle interface would cover if forming a dense relaxed raft. This means that the large majority of the particles stored in the folds (at least 80%) cannot be made available to supply the neighboring surface initially free of particles. From a practical point of view, the self-healing capacity of such interfaces appear very limited under the present conditions.

Let us now consider the raft compressed to the same level but from the front side. The evolution of the unjammed, folded and escaped areas appear to be totally different, see Fig. 3(b). About 60% of the initially confined area unjams. The folded area, which represents at maximum 15% of the confined area totally disappears. Here as well, the evolution of these two quantities seem to be correlated as indicated by the coincidence of the local minimums of $A_{uj}$ with the local maximums of $A_f$, found around 1.4s and 2.2s. Yet, in contrast to back compressed raft, these events do not correspond to the migration, expansion and disintegration of individual folds but to the elimination of two larger folded blocks, found on both sides of the orifice. The first elimination takes place between the third ($t_{f,3} = 0.63\, s$) and fourth ($t_{f,4} = 1.6\, s$) pictures of Fig. 2 while the second one occurs between the fourth ($t_{f,4} = 1.6\, s$) and fifth ($t_{f,5} = 2.8\, s$) pictures. Beside these two bumps, $A_{uj}$ continuously increases while $A_f$ decreases. Almost all particles initially stored in folds unjam and migrate through the orifice to cover a surface that is 1.3 times the excessive surface given by $A_e - A_c$. While a value larger than 1 can first be surprising, it is well explained by the fact that the assembly of escaped particles is less dense than the relaxed raft. The relaxed raft density is close to the one of jamming, defined as $\phi_j = \pi/2\sqrt{3} \approx 0.91$. The one of the escaped assembly can be only roughly estimated from our pictures, providing $\phi_e \approx 0.74$, in agreement with 0.76, the value required to obtain "particle surface conservation" under total unjamming and given by $\phi_e = \phi_j/A_c$.

These results clearly show that if the compression direction is favorable, the self-healing capacity of particle laden interfaces can become very important and approach its theoretical maximum. This maximum is found when all particles in the folds (at least 80%) cannot be made available to supply the neighboring surface initially free of particles. From a practical point of view, the self-healing capacity of such interfaces appear very limited under the present conditions.

At this stage, it is difficult to draw conclusions about the self-healing dynamics. For front compressed raft, only 0.12s are required to obtain $A_e = 15\%$ by comparison to 0.33s for back compressed raft. Yet, this rate quickly slows down. If calculated over a longer period of time (2s), it is found to be $A_e = 0.5\, s^{-1}$, comparable to $0.45\, s^{-1}$, the rather constant rate obtained for back compression.

To better understand the relaxation dynamics, we perform PIV on the unjammed areas found inside and outside the initially confined domain. Representative snapshots of the results are shown in Fig. 4 For the back compressed raft, both the velocity magnitude and direction are uniform. The vectors clearly indicate a flow in the compression direction, from the back of the raft to the orifice and beyond. In contrast, the front compression gives rise to local acceleration, especially close to the orifice, and components perpendicular to the compression direction develop. Interestingly, the velocity field of the unjamming particles inside the confined domain is not symmetric. Indeed, the instant chosen here (1.28s) corresponds to the beginning of the already mentioned elimination of one of the two folded blocks found on each side of the orifice (here, up on the picture).

![Image](image.png)

Figure 4: Exemplary results of the PIV for (a) back compressed raft, $t_b = 0.28\, s$ and (b) front compressed raft, $t_f = 1.28\, s$.

To visualize the PIV results obtained over the whole process, it may be useful to compare the velocity distribution obtained on the sub-phases defined in section 3.3. They are shown in Fig. 5 left and right, for back and front compressed...
Figure 5: Normalized velocity distributions for left: back compressed raft, and right: front compressed raft. Red: unjammed area inside the confined domain; blue: escaped particles outside of it. From top to bottom, the sub-phases are: \( T_{b,1}, T_{b,2}, T_{b,3}, T_{b,4} \) for the back compressed raft; and \( T_{f,1}, T_{f,2}, T_{f,3}, T_{f,4} \) for the front compressed raft. The contours correspond to 0.1, 0.5 and 0.9 of the maximum value.

rafi, respectively. For the back compressed raft, the velocity fields inside (red) and outside (blue) of the confined domain are always very similar, confirming that the unjammed particles form a cohesive assembly, which moves as a block. The velocity component perpendicular to the compression axis, \( v_{\perp} \), is centered in zero and shows very small fluctuations, the 50% contour being comprised in \( \pm 0.01 \text{m/s} \). This is not the case of the parallel component, \( v_{\parallel} \), which has positive values along the whole process. The flow from the back to the front shows some variations with a maximum during \( T_{b,2} \), i.e. when the first fold disintegrates. Its mean value (0.07 m/s) and the fluctuations around it (−0.03 m/s and +0.07 m/s) then decrease until the end of the process.

The velocity fields that develop for the front compressed raft are very different, see Fig. 5. The correspondence between inside and outside is lost, which may indicate a certain independence of the two flows. Inside the confined domain (red), significant movements develop perpendicular to the compression axis leading to \( v_{\perp} \) values as large as \( v_{\parallel} \) values. The flow is not symmetric across the compression axis, see for example \( T_{f,3} \) or \( T_{f,4} \) for which \(-0.01 < v_{\perp} < +0.11 \text{m/s} \) and \(-0.06 < v_{\perp} < 0.01 \text{m/s} \), respectively. This profound asymmetry corresponds to the successive dislocation of two large folded blocks found on each side of the orifice. While passing through the orifice, the particles recover a symmetric velocity field (see blue maps). One can suppose that the shear they experience at this point overcome the capillary attraction, forcing them to rearrange and explaining the reduced density. Finally, the focus of the velocities toward zero can rather be attributed to a normalizing effect than to changes in the flow close to the orifice. The increasing investigated surface area \( (A_e) \) contains an increasing number of points with low velocity, typically located at the assembly periphery, and therefore focuses the normalized distribution.

Despite the important disparities of the velocity fields of the front and back compressed rafts, the velocity profiles at the orifice seem first rather similar. The profiles displayed in Fig. 6 do not evidence obvious differences. The shape of the profile, the value of the maximum velocity and the importance of the fluctuations are comparable in both cases.

To better analyse the particle flux and its fluctuations, we now focus on the mean velocity \( \overline{u} \). The results are plotted in Fig. 7 together with \( \Pi \), the stress measured at the raft back. Two points are worth being discussed. First, the different evolution of \( \overline{u} \) for the front and back compressed rafts. For the back compression, after a short transitory phase and before the relaxation stops, \( \overline{u} \) seems rather constant. The two peaks, attributed to the disintegration of two successive folds (see Fig. 2), can be identified but no clear increase or decrease of \( \overline{u} \) over time can be seen. It is not the case for the front compressed raft for which \( \overline{u} \) clearly decreases over time. Fluctuations are perturbing this evolution but they can be well explained by the already mentioned elimination of the two folded blocks found on each side of the orifice. This confirms that the compression side does not only influence the self-healing capacity but also its kinetics. The second
important point evidenced by Fig. 7 concerns the stress measured at the back of the raft. The latter remains almost unchanged for the back compressed raft, indicating that an important fraction of the initially applied stress remains. This remaining stress most likely originates the still compressed portions found on each side of the central unjammed corridor. A closer look at the curve indicates a slight and probably step-wise decrease of $\Pi$ after each fold disintegration. If confirmed, such variations are typical of a solid-like behavior, for which the stress and strain are linearly connected via the Young modulus. For the front compressed raft, the stress totally relaxes. Interestingly, fluctuations are observed that correspond to the ones of $u$, indicating a direct and almost instantaneous transmission of the stress trough the entire raft. This could be interpreted as a liquid-like behavior, for which the stress and strain rate are proportional, the proportionality coefficient being the viscosity.

### 4.2 Generalisation and interpretation

The detailed results presented above were obtained on two similar rafts and generalizing these findings requires more data. Thus, these experiments were repeated with two other pairs of raft, compressed either from the front or from the back, with a compression level of 33% and 50%, respectively.

The final relaxation states are characterized in Fig. 8 and appear to be mostly fixed by the compression side. For rafts compressed on the back side (left picture raw), the relaxation is partial, folds remain and the escaped particles form a rather dense assembly, whose width is the orifice width. Similar rafts compressed from the front side give rise to a quite different outcome, see right picture raw. The relaxation is almost total, only a few folds remain if any, and the escaped particles form less dense assemblies whose width is much greater than the orifice width. A more quantitative description is shown in the central plots where $A_e$ (squares), $A_{uj}$ (diamonds) and $A_f$ (triangles) are reported as a function of $K$. Whatever the level of compression, the relaxation of the front compressed rafts (full symbols) is always much more complete than the one of the back compressed rafts (empty symbols). In other words, $A_e$ and $A_{uj}$ are systematically greater for front compression (factor 4 to 8 and 3 to 8, respectively) while the opposite is observed for $A_f$. Focusing on the back compressed rafts, $K$ has a limited influence on $A_e$ and $A_{uj}$ but strong effects on $A_f$. For front compression, the influence of $K$ on $A_e$ and $A_{uj}$ is significant. The greater the compression, the greater these two quantities. The effect is stronger for $A_e$ than $A_{uj}$, which could be explained by variations of the assembly density. Indeed, the surface occupied by a given number of escaped particles is modulated by the packing density of the assembly, which possibly
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Figure 8: Plots (central raw): final values of $A_e$, $A_{uj}$ and $A_f$ as a function of $K$ with empty (full) symbols for back (front) compression. Pictures: final relaxation states obtained for back (left raw) and front (right raw) compressed rafts. From top to bottom, the raft compression is: $K=$33%, 50% and 67%.

decreases with increasing compression (or flow velocity). Folds are very limited, which is expected given the large scale unjamming.

From a practical point of view, these results indicate that for the studied geometry, the self-healing capacity of particle laden interfaces is principally a function of the compression direction. For a given direction, the greater the compression, the greater the fraction of stored particles that can flow and cover initially particle free regions. This released fraction is always much larger for front than for back compression.

To go further and better assess the potential of folds as particle reservoirs, the dynamics of this release should be characterized. We therefore plot in Fig. 9 the temporal evolution of $A_e$ for all studied rafts. The back compression does not only limit the magnitude of the particle release but also its duration. The latter is always less than 0.5s, to be compared with 2s to 4s for similar compression achieved from the front side. Said differently, the incomplete character of the relaxation could, at first glance, be explained by a premature arrest of the process. Yet, a more detailed analysis of our data points toward more complex phenomenon.

Let us start considering the front compressed rafts, see Fig. 9(b). For interpreting the release dynamics, it may be useful to make an analogy with an electric system. In this frame, the instantaneous rate of release, or flow, $-\varphi_e A_e$, corresponds to the current $i$. The flow is subjected to some resistance, $R$, which we assume to be constant. The confined area can be modeled as a capacitor of capacitance $C$, whose charge $q$ corresponds, at first order, to the excess of particles. Thus, for any instant $t$, $q = \varphi_j(A_r - A_e) - \varphi_e A_e$, where $\varphi_j(A_r - A_e)$ represents the initial excess of particles and $\varphi_e A_e$ the escaped particles. Noting $U$ the potential difference, the capacitor discharge through a resistor $R$ is given by

$$C \frac{dU}{dt} + \frac{U}{R} = 0$$

(1)

This equation can be solved for given initial conditions and provide $U(t)$, and by extension $i(t) = U(t)/R$. 

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Figure 9: $A_e(t)$ for (a) back and (b) front compressed rafts. Symbols are experimental data with triangles for $K = 33\%$, diamonds for $K = 50\%$ and circles for $K = 67\%$. The dashed and continuous lines are linear and exponential fits given by Eq. (3) and (4), respectively. Insets: $A_e(t = 0)$ (squares) and $\tau$ (stars, if applicable) as a function of $K$.

In the present system, the first term of Eq. (1), $CdU/dt = dq/dt = i$, corresponds to $-\varphi_e \dot{A}_e$ and the second one, $U/R = q/RC$, to $[(\varphi_j(A_r - A_c) - \varphi_e A_e)]/RC$. The discharge equation therefore becomes

$$\dot{A}_e + \frac{A_e - (A_r - A_c)\varphi_j/\varphi_e}{RC} = 0$$

(2)

and the solution verifying the initial condition $A_e(t = 0) = 0$ is then:

$$A_e = A_0(1 - e^{-t/\tau})$$

(3)

with $A_0 = (A_r - A_c)\varphi_j/\varphi_e$, the final escaped surface and $\tau = RC$, the typical release time constant.

To evaluate the relevance of our analogy, the experimental evolution of $A_e$ is fitted by Eq. (3) letting $A_0$ and $\tau$ be adjusted to minimize the sum of square residuals. The agreement is excellent, see Fig. 9(b). The biggest deviations are indeed observed either at the very beginning or at the end. The former is attributed to initial perturbations caused for example by capillary wave, by a possible short transient regime or by the difficulty to precisely identify the time origin. The discrepancy found in the last instants may be caused by a premature arrest of the relaxation, which is discussed later. The values of $\tau$ and $A_0/\tau$ produced by the fitting procedure are plotted in the inset as stars and squares, respectively. Both quantities seem to increase roughly linearly with $K$. Given the limited number of points, the interpretation remains tentative but one could postulate that $R$, the resistance to the flow, is similar for all (front compressed) rafts, while $C$, the system capacitance, is directly proportional to the amount of particles stored in the folds, and thus to $K$. The initial discharge rate given by $A_0/\tau$ also increases with $K$, but the linear character is less pronounced. If confirmed, it would indicate that $\varphi_j/\varphi_e$ varies as $K^2$. Future experiments could be used to probe this scaling, the variations of $\varphi_e$ being important in view of using folds as particles reservoir.

Let us now focus on back compressed rafts. The experimental points of Fig. 9(a) cannot be immediately identified with an exponential decay, which questions the previous interpretation. The latter considers the emptying of a reservoir subjected to a pressure, which decreases linearly with $A_e$, and to losses, which are proportional to the flow, i.e. scaling as $\dot{A}_e$. Given the very limited decrease of $\Pi_0$ observed during the relaxation of back compressed rafts, it is legitimate to consider a constant pressure. Keeping the rest unchanged leads to a simple linear increase of $A_e$ with $t$, which reads:

$$A_e = \frac{\Pi_0}{\varphi_e R} t$$

(4)

Here $\Pi_0$ is the constant pressure applied at the back of the raft, $\varphi_e$ and $R$ are unchanged and represent the escaped particle density and the resistance to the flow, respectively. To test this model, the experimental results are fitted by linear functions, see dashed lines in Fig. 9(a). Note that not zero intercepts are enabled since the experimental curves show an initial step. Beside this, the agreement is reasonable with the largest deviations observed at the end of the process. Interestingly, assuming that $\Pi_0 \propto A_r - A_c$, we expect that the slope $\Pi_0/(\varphi_e R)$ of fixed size rafts (constant $A_r$) is proportional to $K$. This is indeed in good agreement with our data, see inset of Fig. 9(a), which shows - despite the limited number of points - a linear variation of the fitted slopes with $K$. These curves can therefore be seen as classical
emptying of granular silos. Indeed, the well known Beverloo law predicts for given particle and orifice sizes, a constant
flow rate [Beverloo et al. (1961)]. The origin of this law remains controversial and neither the Janssen effect [Janssen
(1895)] nor the free falling arch approach seem to provide the correct view [Rubio-Largo et al. (2015)]. Furthermore, the
dependency of the flow rate \((A_c)\) with the pressure remains unclear. Some experiments evidence a total independence
[Aguirre et al. (2010)], while others show - under certain circumstances - a proportional relation [Peng et al. (2021)], in
agreement with our findings. Yet, it is worth noting that these results, i.e. \(A_c(t) \propto t\) and \(A_c \propto K\), are also compatible
with the premature arrest of exponential release as found for front compressed rafts. One must here keep in mind that
the limited character of the current data does allow to choose one of the two models.

Whatever the chosen function, the question of why and when the relaxation process get arrested remains open. We
attribute the ending to the existence of a yield stress, at least for back compressed rafts. Thus as long as the pressure \(\Pi\)
is above certain critical value, flow can occur. Small decrease of \(\Pi\), even limited and at first order negligible (hypothesis
of constant \(\Pi_0\)), can then stop the flow. Coming back to the electrical analogy, everything happens as if compressed
rafts were granular diodes. When the diode is mounted in the appropriate direction, here corresponding to the front
compression, the capacitor constituted by the particles stored in the folds can (quasi)-totally discharge, the current
passing through a constant resistor \(R\). Yet, if the compression occurs from the back, the diode, mounted in the opposite
direction, stops - after some leaking current has passed - the capacitor discharge.

In the context of compressed particle-laden interfaces, this diode effect can be understood in the light of granular
framework, which describes the development of force chain network during the compression. By moving one barrier
while keeping the other fixed, chains build up starting from the moving barrier and propagate via particle-particle
contacts. For some of these contacts, chains divide forming two or more branches. Considering the monodisperse
character of the particles, and by extension their hexagonal close packing, the orientation of these chains and branches
are expected to be found in a cone, whose axis is the compression axis and angle is \(60^\circ\). Consequently, for rafts
compressed from the back side, the network transmits the stress toward the front barrier but an important portion of it
is redirected on each side of the orifice. The keystones, i.e. particles for which the chains branch, are in the back of
the raft and therefore not easily removed. Arches may form that block the unjamming, which only occurs in a narrow
corridor whose width is more or less the orifice width. In contrast, when rafts are compressed from the front, the gate
opening causes the removal of keystone particles found right behind it, leading the force chain network to collapse, and
therefore triggering a quasi-total unjamming of the confined area. A careful inspection of the unjammed zone observed
at the beginning of the process (see for example Fig. 2(2)\(f_{f,2}\) let see a conical shape whose angle is very close to \(60^\circ\),
in perfect agreement with the expected network structure. Finally, this interpretation also explains why under certain
circumstances, such as very strong compressions, also front compressed rafts may give rise to a force chain structure
that can, at least partially, arrest the unjamming, see Fig. \(K = 67\%\) (circles) in Fig. 9(b).

5 Conclusions

The different behaviors observed during the relaxation of strongly compressed rafts evidence the importance of the
compression direction on these systems. For back compressed rafts, the relaxation is incomplete while it is quasi-total
for front compressed rafts. Practically, these results demonstrate that the processability of these interfaces highly
depends on their history. In the present configuration, the flowability is primarily set by the compression direction, the
level of compression playing a secondary role. This important finding should be accounted for in industrial processes.
It is also of importance while considering the self-healing properties of these interfaces. Indeed, in this simple uni-axial
geometry, the capacity of particles stored into large folds to migrate and stabilize uncovered areas is almost total for
favorable compression direction, while it is strongly hindered in the opposite case. The dynamics of the particle release
itself is strongly affected by the compression direction. While the current data remain limited, we observe similar trends
in on-going tests performed using different raft size, orifice width and particle size.

These results are interpreted in the light of the framework developed for granular matter, which demonstrates the
importance of chain forces to transmit stress. Thus, the direction of these chains, and more precisely their branching, is
found to be essential as evidenced by triggering a local relaxation along the compression axis. For front compressed
rafts, the local unjamming causes keystone particles to be removed leading to the network collapse and large scale
unjamming. In contrast, for back compressed rafts the network redirects the stress laterally. As a result, only a limited
number of particles found directly behind the orifice can flow, limiting the relaxation to a narrow corridor. To visualize
these effects, the analogy with an electrical circuit can be made. The folds are like a capacitor and the particle flow like
a current through a resistor. The force chain network can then be seen as a diode which let, or not, the current flow
depending on its orientation.

Finally, it is worth noting that our results and interpretation give rise to many open questions which need to be treated in
future research. From our point of view, future investigations should aim in understanding what influences the properties
of the chain force network. In this frame, we can of course think of the raft and trough geometry, and especially of the raft length, its width and the size of the orifice. A second important aspect is the influence of the individual particles, and more particularly, the role of friction, shape, but also possible contact lubrication, which could for example cause network aging.

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Appendix

This Appendix presents 1) the preparation of the elastic barrier, 2) the method used to calibrate it and 3) the deformation equations, which are then used to deduce the lineic pressure acting on it.

1) Elastic rubber preparation

The elastic rubber strings are produced in house by injecting a freshly prepared (1:1) mixture of Zhermack Elite Double 8 basis and catalyst (Zhermac Spa) into glass capillaries. After the elastomer reticulation has been completed, the glass capillaries are manually removed. Using capillaries of various diameter, we produce strings whose diameter ranges between 0.5 and 1 mm. Their typical length is in the range of 10 cm, which is sufficient to be installed in 6 cm wide trough.

2) Rubber calibration

The calibration of the rubber mostly consists in determining its Young’s modulus. The principle, detailed below, is simple, and consists in measuring the deflections produced by hanging known weights to the rubber, see Fig. 10. It was first presented in the PhD thesis of Petit (2014).

Consider an elastic string of relaxed length \( s_0 \) (not precisely known), attached between two holders separated by \( s_1 \) (black rectangles in Fig. 10). The elastic is further loaded using a known mass \( m \) \( (F_g = mg) \) and reaches a stretched length \( s_f \), which can easily be measured. The tensile stress, \( \sigma \), is linearly proportional to the strain, \( \epsilon \), and the proportionality coefficient, \( E \), is the Young’s modulus. This reads:

\[
\sigma = E \epsilon \quad (5)
\]

The strain is equal to the relative extension and the tensile stress derives from the tension \( T \), providing:

\[
\epsilon = \frac{s_f - s_0}{s_0} \quad \text{and} \quad \sigma = \frac{T}{\Sigma} \quad (6)
\]

Figure 10: Principle of the elastic calibration: deformation is measured as a function of the applied force.
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with Σ, the cross-section area of the elastic string under tension. The Hook’s law may now be rewritten as

\[ T = E \Sigma \frac{s_f - s_0}{s_0} = E \Sigma_0 \frac{s_f - s_0}{s_f} \]  

(7)

where the variation of the cross-section area due to the stretching was introduced as

\[ \Sigma = \Sigma_0 \frac{s_0}{s_f} \]  

(8)

Σ₀ is the cross-section area of the non-deformed elastic string. Under a small load, the force equilibrium may be written as:

\[ F_g = 2T \sin \beta \quad \text{with} \quad \sin \beta = \sqrt{1 - \left( \frac{s_1}{s_f} \right)^2} \]  

(9)

Here, β is the angle between the rubber string and the horizontal. Combining the above equations, one obtains the following relationship:

\[ mg = 2\lambda_0 \cdot \frac{s_f - s_0}{s_f} \sqrt{1 - \left( \frac{s_1}{s_f} \right)^2} \]  

(10)

with the newly introduced parameter λ₀, defined by \( \lambda_0 = E\Sigma_0 \).

Practically, the length of the deformed rubber \( s_f \), is measured from pictures taken for different known masses \( m \). The undeformed rubber length, \( s_0 \), which is not precisely known and the parameter \( \lambda_0 \) are then obtained by finding the best fit to the measured data. This is done with the help of the \textit{fminsearch} function of Matlab. An illustrative curve is displayed in Fig. 11. The coefficient of regression is always very close to one, greater than 0.99. The 95% confident intervals corresponds to an uncertainty of ±1% on \( \lambda_0 \) and \( s_0 \). Knowing \( \Sigma_0 \), the cross section of the undeformed string,

![Figure 11: Typical calibration curve and its linear fit providing \( s_0 \) (here 44.46 mm) and \( \lambda_0 \) (here 0.3152 N). Function \( f(s_f) \) is defined as \( f(s_f) = 2 \frac{s_f - s_0}{s_0} \sqrt{1 - \left( \frac{s_1}{s_f} \right)^2} \). The coefficient of regression is \( R^2 = 0.9995 \).](image)

the modulus of elasticity is finally deduced as:

\[ E = \frac{\lambda_0}{\Sigma_0} \]  

(11)

Using the Elite Double 8, the measured \( E \) is found to be in the expected range of 0.1 MPa. Note, that the two parameters \( s_0 \) and \( \lambda_0 \) are also necessary to describe the deformation of the elastic string subjected to a constant lineic force, as shown in the next section.

3) Deformation equations

The calibrated rubber string can then be used as a pressure sensor for the particle rafts in our experiments. To be quantitative, the relationship between the elastic deflection, \( \delta \), and the applied lineic pressure, \( \Pi \), is needed. These quantities are defined in Fig. 12. The length of the stretched elastic is \( s_f \), as previously defined. The position along the
elastic can be described by the curvilinear ordinate \( s \) or by the cartesian coordinates \((x, y)\), the \( x-\) and \( y-\)axes have unit vectors \( \hat{x} \) and \( \hat{y} \), respectively.

The following derivation is based on the well-known mathematical problem of a classical catenary [Borggräfe et al. (2015)]. The tension \( \vec{T} \) has a norm \( (T) \) and is collinear to the string at any point \( s \in [-s_f/2; s_f/2] \). Thus, naming \( T_x \) and \( T_y \) its horizontal and vertical components, we get:

\[
\frac{dy}{dx} = \frac{T_y}{T_x} \quad \text{and} \quad T = \sqrt{T_x^2 + T_y^2} \quad (12)
\]

Using the definition of the lineic pressure, \( \Pi \), and applying the force balance to a string element of infinitesimal length \( ds \) (see Fig. 12) provides:

\[
T_y = \int_{0}^{s_f/2} \Pi ds = \Pi \int_{0}^{s_f/2} ds \quad (13)
\]

which leads with Eq. (12) to:

\[
\frac{dy}{dx} = \frac{\Pi}{T_x} \int_{0}^{s_f/2} ds = \frac{\Pi}{T_x} \int_{0}^{s_f/2} \frac{ds}{dx} dx
\]

Eq. (14) can be differentiated with respect to \( x \), leading to:

\[
\frac{d^2y}{dx^2} = \frac{\Pi}{T_x} \frac{ds}{dx} \quad (15)
\]

To go further, the curvilinear ordinate is eliminated using:

\[
ds^2 = dx^2 + dy^2 = dx^2 \left( 1 + \left( \frac{dy}{dx} \right)^2 \right) \quad (16)
\]

which also writes:

\[
\frac{ds}{dx} = \sqrt{1 + \left( \frac{dy}{dx} \right)^2} \quad (17)
\]

Eq. (15) becomes:

\[
\frac{d^2y}{dx^2} = \frac{\Pi}{T_x} \sqrt{1 + \left( \frac{dy}{dx} \right)^2} \quad (18)
\]

It is integrated with boundary conditions \( dy/dx(x = 0) = 0 \) and \( y(x = 0) = 0 \), providing:

\[
y(x) = \frac{T_x}{\Pi} \left( \cosh \left( \frac{\Pi s_1}{2T_x} \right) - 1 \right) \quad (19)
\]

Evaluating this expression for \( x = s_1/2 \) gives the central string deflection:

\[
\delta = \frac{T_x}{\Pi} \left( \cosh \left( \frac{\Pi s_1}{2T_x} \right) - 1 \right) \quad (20)
\]

In practise, to use this expression, it is necessary to explicit \( T_x \) as a function of the known parameters, i.e. \( s_1 \), \( s_0 \) and \( \lambda_0 \). Combining Eq. (12) and (13), we get:

\[
T = \sqrt{T_x^2 + \left( \Pi \int_{0}^{s_f/2} ds \right)^2} \quad (21)
\]
Using the stress-strain relation from the calibration section (Eq. (7)) and the definition of the parameter $\lambda_0$ given by Eq. (11), we identify $T$ with:

$$T = \lambda_0 \frac{s_f - s_0}{s_f}$$

and obtain:

$$\lambda_0 \frac{s_f - s_0}{s_f} = \sqrt{T_x^2 + \left( \frac{\Pi}{T_x} \int_0^{s_f/2} ds \right)^2}$$

We then eliminate $s_f$ using:

$$s_f = \int_0^{s_f/2} ds = \int_0^{s_1/2} \sqrt{1 + \left( \frac{dy}{dx} \right)^2} dx = \frac{T_x}{\Pi} \sinh \left( \frac{\Pi s_1}{2T_x} \right)$$

Eq. (23) and (24) provide the condition

$$\lambda_0 \frac{T_x}{\Pi} \sinh \left( \frac{\Pi s_1}{2T_x} \right) - \frac{s_0}{s_f} = \sqrt{T_x^2 \left( 1 + \sinh^2 \left( \frac{\Pi s_1}{2T_x} \right) \right)}$$

or equivalently

$$T_x \cosh \left( \frac{\Pi s_1}{2T_x} \right) = \lambda_0 \left( 1 - \frac{s_0 \Pi}{2T_x \sinh \left( \frac{\Pi s_1}{2T_x} \right)} \right)$$

The relation between $\delta$, the central deflection of the elastic string and $\Pi$, the applied lineic pressure is therefore given by the two equations (20) and (26). Note that, using Eq. (19) and (26), the entire shape of the elastic is indeed described as a function of known and measurable parameters, namely $s_1$, $s_0$ and $\lambda_0$.

For small deformations, Eq. (20) and (26) can be simplified by a third-order series expansion with the approximation

$$\frac{dy}{dx} \approx \frac{s_1 \Pi}{T_x}$$

which provides the following relation between the lineic pressure $\Pi$ and the (small) central deflection $\delta$:

$$\Pi = \frac{8 \lambda_0}{s_1} \left[ \left( 1 - \frac{s_0}{s_1} \right) \frac{\delta}{s_1} + \left( \frac{4s_0}{3s_1} - 1 \right) \left( \frac{2\delta}{s_1} \right)^3 \right]$$

When applying the lineic pressure measurement method described above, a non-negligible uncertainty must be taken into account. The accuracy depends strongly on the difference $s_1 - s_0$ as shown below:

$$\frac{\Delta \Pi}{\Pi} = 2 \frac{\Delta s_1}{s_1} + \frac{s_1 \Delta s_0 + s_0 \Delta s_1}{s_1(s_1 - s_0)}$$

In the present work, we evaluate this uncertainty to approximately 15%.

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