Structural properties of crumpled cream layers

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Received 9 April 2007, in final form 10 April 2007
Published 4 June 2007
Online at stacks.iop.org/JPhysD/40/3665

Abstract
The cream layer is a complex heterogeneous material of biological origin which forms spontaneously at the air–milk interface. Here, the crumpling of a single cream layer packing under its own weight at room temperature in a three-dimensional space is studied. The structure obtained in these circumstances has a low volume fraction and anomalous fractal dimensions. Direct means and noninvasive NMR imaging techniques are used to investigate the internal and the external structures of these systems.

1. Introduction

The crumpling of a surface, a sheet polymer or a self-avoiding elastic model sheet, is a subject of great interest in both theoretical [1, 2], and applied physics [3, 4], as well as in materials science [5], due to the peculiar properties of the crumpled structures. It is known that both a paper sheet and a thin metal foil of area $A$ and mass $M$, when submitted to a haphazard ill-defined compaction by manual means in order to generate an approximately globular object, present a non-thermal transition to a crumpled fractal state of packing characterized by the formation of a complex pattern of folds. The crumpled structures obtained with these materials obey the nontrivial scaling

$$A \sim M \sim \phi^{D_M},$$

where $\phi$ is the corresponding average globular diameter of the structure after crumpling and $D_M$ is the mass fractal dimension [3, 6, 7]. This fractal dimension assumes in these cases values in the interval $2.2 < D_M < 2.7$ irrespective of the material and of the thickness of the foil in the studied interval of 20–200 $\mu$m [3, 6, 7].

Anyone knows from childhood that from ordinary milk we can obtain a cream layer, that tenuous whitish membranous layer which is formed on the upper part of the cream right at the air–milk interface. The cream layer obtained from milk is a thin sheet that contains micrometre- and submicrometre-sized fat globules, proteins, phospholipids and water, among other components, and is formed very rapidly, within a few minutes, in cold milk. The cream layer is formed at the free surface of milk as a planar quasi-two-dimensional continuous internal fat network [8, 9] as a result of a complex process of coalescence of fat globules. Milk is in fact one of the most complex foods, with more than 100 000 different molecular components [8]. Besides its economic importance in dairy industry, milk is the most important source of nourishment and immunological protection for young mammals, and for humans it has been a food source since prehistoric times.

Here we investigate the crumpling associated with the three-dimensional packing of a single cream layer. Our extensive analysis indicates that the cream layer collapses under its own weight at room temperature into a three-dimensional fractal structure of low volume fraction. The structure of this paper is the following: in section 2 we describe the experimental details, in section 3 we present our results and in section 4 we have a summary of our conclusions.
made at the room temperature of excess liquid is lost. All the measurements reported here were in the range of 30–40 s. This period includes a stage of drainage in which all transference and deposition of each cream layer is typically completed and studied. The total time involved in the stages of transference from the milk-free surface to the surface where it is deposited and studied. In the present study, using direct means and noninvasive imaging techniques, we study the crumpled state of sample paper which is observed when a cream layer is slowly deposited onto a glass support and packs fractally under the action of its own weight. In this case (CCL#1), the original cream layer had an area of 638 cm² and an average thickness of 1 cm. The packing process is arrested in a fractal state of low density and the radius of the planar fresh cream layer. Figure 1(b) shows the external appearance of the cream layer after crumpling (CCL#1). The three-dimensional diameter of the CCL#1 was 3.1 cm and the volume fraction η ≃ 0.33, i.e. approximately 51% of the volume fraction for random close packing of spheres in three-dimensions [11]. For comparison, the photograph of a crumpled surface of paper of a similar external size and a thickness of 125 µm is shown in figure 1(c).

3. Results and discussion

There are various ways to characterize the basic structural properties of real irregular surfaces [12, 13]. Here, we choose to investigate the geometric properties of CCLs in terms of both the mass fractal dimension and the box dimension. Firstly, we measured the dependence of the area A with the average three-dimensional diameter φ (equation (1)) for the ensemble of 72 CCLs. The experimental samples of cream layers studied here had an area (average diameter) in the interval 3 ⩽ A(cm²) ⩽ 638 (0.4 ⩽ φ(cm) ⩽ 4.3) and a surface thickness ζ = (82 ± 9) µm. The A × φ measurements give rise to the plots shown in figures 2(a) and (b), respectively, for fresh (f) samples as well as for dry (d) rigid samples examined 10 days after CCL formation, and consequently with low water content. The continuous lines in these plots refer to the corresponding best fits A ∼ φ^0.15±0.10 and A ∼ φ^2.65±0.10, i.e. there is a slight increase in the mass fractal dimension (and a slight reduction in the statistical fluctuations) with the ageing of the system, that is when the system evolves from fresh CCL to dry CCL. The external measurements of the diameter of the CCLs suggest that after crumpling these systems condense in the three-dimensional physical space as an anomalous non-space-filling structure with mass fractal dimension D_M < 3. However, the external measurements of diameter and area are
due to the $T_1$ relaxation process. Mostly, we have used an imaging matrix of $128 \times 64 \times 64$, slice thickness of 1 mm, and $T_R \geq 1.2$ ms. Each three-dimensional image took 68 min. Since experiments are short in time, no appreciable loss of liquid by the samples is observed. After all corrections we can state that pixel intensities on the images are proportional to free and almost-free water content and to protons in small molecules. Finally, it is possible that NMR images might not represent the total mass distribution of CCLs exactly. If this is the case, our estimate for the ensemble average of the box fractal dimension for CCLs could be somewhat underestimated.

Thus, images of two-dimensional sections of large fresh CCLs were obtained along three orthogonal planes $xy$, $yz$ (both in the direction of the gravitational field) and $xz$ (orthogonal to the gravitational field) and in intervals separated by a distance of 1 mm. Figures 3(a) and (b) ((c) and (d)) show the NMR images (and the corresponding box-counting analysis [16] that gives the number $N(\epsilon)$ of square boxes of size $\epsilon \times \epsilon$ needed to cover the image) along, respectively, some intermediary planes $xy$ and $xz$ for CCL#1, for three different values of the experimental tuning parameters. The images refer to the situation of the absence of any filtering on the signal ($F = 0$) and with a filtering factor of 25% ($F = 0.25$). In figures 3(a) and (b) each one of the three columns refer to a particular set of experimental tuning parameters. Both box-counting plots are well described by the scaling $N(\epsilon) \sim \epsilon^{-D}$ along 1.5 decades in $\epsilon$, as can be seen from the power law best fits (continuous straight lines in figures 3(c) and (d)), both with a slope of $\delta = 1.30 \pm 0.05$. This value of $\delta$ represents the fractal dimension of those particular sections [16].

The corresponding box fractal dimension of the CCL in a three-dimensional physical space [16] is $D = \delta + 1 = 2.30 \pm 0.05$. The fluctuation bars in figures 3(c) and (d) are associated with the means on different image planes and experimental tuning parameters. In general, the value of $D$ for any sample is robust within typical statistical fluctuations of 3–5%, irrespective of the value of $F$ and the other experimental parameters.

In figure 4 we show the box-counting plot for the entire ensemble of CCL images studied: 550 experimental data points for $F = 0.25$, after averaging on image samples, on three orthogonal image planes ($xy$, $yz$ and $xz$) and on experimental tuning parameters. The slopes of the straight lines associated with the power law fits are $\delta_{xy} = 1.34 \pm 0.03$, for 439 images with $64^2$ pixels (dashed line) from CCL#1, and $\delta_{yz} = 1.53 \pm 0.07$, for 111 images with $426^2$ pixels (continuous line) from CCL#2 ($A = 625 \text{ cm}^2$, $\phi = 3.0 \text{ cm}$, and $\eta \simeq 0.36$). CCL#1 and CCL#2 were the two largest samples in the ensemble studied. The fluctuations bars are due to the mean on many orthogonal image planes and on experimental tuning parameters. The weighted ensemble mean of these values gives the result $\delta_{ens} = 1.38 \pm 0.10$. No noticeable change in $\delta_{ens}$ was observed within practically the full interval of variation of $F$. Thus, our overall estimate for the ensemble average of the box fractal dimension of the CCL is $D_{ens} = \delta_{ens} + 1 = 2.38 \pm 0.10$, which is equal within the fluctuation bars to the mass dimension obtained from figure 2(a) and from experiments with crumpled sheets of paper [6,7], metal foils [3], as well as from computer simulations [5] and a Flory-type approximation expected to be a value for membranes at thermal equilibrium [10].
Figure 3. Typical NMR images of cross sections of fresh CCL#1 and box-counting plots: (a) plane xy (parallel to the gravitational field). (b) Plane xz (orthogonal to the gravitational field). The images have 642 pixels and refer to the absence of any filtering on the signal \( F = 0 \) and to a filtering factor of 25% \( (F = 0.25) \). (c) and (d) show the corresponding box-counting plot [16] that gives the number \( N(\epsilon) \) of boxes of size \( \epsilon \times \epsilon \) needed to cover the image (for \( F = 0.25 \)). The plots give the scaling \( N(\epsilon) \sim \epsilon^{-\delta} \) (continuous line), with \( \delta = 1.30 \pm 0.05 \). The fractal dimension of the CCL#1 in a three-dimensional physical space is \( D = \delta + 1 = 2.30 \pm 0.05 \). Fluctuation bars are associated with means on different (parallel) image planes and experimental tuning parameters.

Figure 4. Box-counting plot for large fresh CCLs, for \( F = 0.25 \). The dashed (continuous) line is associated with CCL#1 (CCL#2). Our overall estimate for the box dimension of the ensemble of CCL is \( D_{\text{ens}} = 1 + \delta_{\text{ens}} = 2.38 \pm 0.10 \).

4. Conclusions

Our results based on a large number of experiments of deposition of thin cream layers of cow milk onto a glass support have shown that, although the cream layers collapse under their own weight at room temperature, they never collapse into a three-dimensional compact structure. Rather, the packing process of these tenuous CCL structures is arrested in an intermediary crumpled state of low volume fraction. This crumpled state is surprisingly rigid in the case of dry samples. Two important experimental aspects of this low-density state are its anomalous area-size scaling and its anomalous box dimension. To the best of our knowledge, it is the first time that noninvasive NMR imaging is used to obtain information on the interior of crumpled surfaces (figures 3 and 4). Moreover, the measured fractal dimension of a CCL, a system of animal origin, is equal to that observed for other crumpled surfaces made of completely different materials and obtained by completely different means [3,6,7]. These findings suggest that a universal dynamics may be responsible for all these crumpling processes. In particular, the results reported here seem to confirm that the crumpling dynamics is heavily dependent on a few attributes of the system, as exemplified by the two-dimensional topology of the surfaces [3]. In conformity with a recent work [17], and perhaps most importantly, our results indicate that to a large extent, the fractal dimension of the crumpled surfaces does not depend on the magnitude of attractive interactions transverse to the layer, which are expected to exist in the case of CCL but are absent for metal foils and sheets of paper. The robustness of the numerical value of the mass fractal dimension observed in the macroscopic crumpling experiments reported here for CCL, and in other works for different materials and conditions [3,6,17], may be an indication that a similar type of anomalous packing can be found when the size of the surfaces is reduced to micrometre\(^2\) area scales.

Acknowledgments

This work was supported in part by Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), Programa
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de Núcleos de Excelência and Núcleo de Materiais Avançados (Brazilian government agencies). CCD acknowledges a postdoctoral fellowship from CNPq. The authors are grateful to E N Azevedo for his technical assistance with part of the NMR images. MAFG expresses his thanks to G L Vasconcelos, and I R Tsang for their fruitful discussions.

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