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An experimental study of columnar crystals using monodisperse microbubbles

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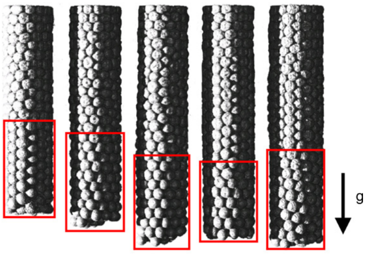
HIGHLIGHTS

- We study the ordering of monodisperse microbubbles within cylindrical confinement.
- X-ray tomography was used to obtain three-dimensional images of their structures.
- Close structural resemblances were found between foam and hard-sphere packings.
- Previously unseen larger structures were formed of several distinct ordered layers.
- A new model predicted the change of contact number with cylinder diameter.

GRAPHICAL ABSTRACT

Reconstruction of the raw three-dimensional tomographic data showing the cylindrical ordering of microbubbles. The red boxes indicate regions of consistent ordering, above which a sharp transition to a different ordered structure is observed. Such columnar crystals have been observed in a wide variety of physical systems.



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ABSTRACT

We investigate the ordered arrangements of monodisperse microbubbles confined within narrow cylinders. These foams were imaged using X-ray tomography, allowing the 3D positions of the bubbles of the foam to be accurately determined. The structure of these foams closely resemble the minimum energy configuration of hard spheres in cylindrical confinement as found in simulations. For larger ratios, λ, of cylinder to bubble diameter two- and three-layered crystals were formed. Each layer of these structures is found to be ordered, with each internal layer resembling structures found at lower λ values. The average number of contacts per bubble is seen to increase with λ.

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1. Introduction

Finding the densest packing of hard spheres is an enduring mathematical problem that finds numerous applications in physics, biology and material sciences. The recent proof of the Kepler conjecture represents a significant milestone in understanding the packing of such objects in an unbounded volume [1]. In contrast, far less is known about packing within a bounded space, despite the fact that such problems are ubiquitous throughout science, nature and even daily life.

A fascinating example of this type of problem is that of finding the densest packing of equal sized spheres (of diameter d) in a cylinder (of internal diameter D). Simulations conducted by us (and

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others) have so far identified some 40 distinct spiral structures in the range $1 < (D/d) < 2.873$ – the majority of which are chiral (with achiral packings arising at particular values of D/d) [2–4]. These structures were identified using *simulated annealing*, a numerical technique which can find the minimum energy configuration of a system [3]. Previously, we have dubbed these helical structures columnar crystals and given a theoretical understanding of some of them [3].

Interest in these columnar crystal is driven by the fact that they are chiral and that the degree of chirality depends directly on the ratio D/d. Consequently, such structures might find numerous opto-electronic applications, as guides to understanding helical formations in biology (e.g. tobacco virus, flagella and microtubules), or as promising pathways to mimic such biological microstructures [5].

In the range $1 < (D/d) < 2.715$ one observes monolayer arrangements (in which all the spheres are in contact with the confining cylinder) while for larger values of D/d we observe more complex multilayer structures that include internal spheres (i.e. spheres which are in contact with other spheres but not in contact with the cylinder). Finding dense hard sphere packings for large values of D/d is a computationally demanding task and as such multilayer arrangements remain largely unexplored [3]. In this paper we show that micron-sized spherical bubbles can spontaneously self-assemble into crystalline arrangements inside capillary tubes, which closely resemble multilayer columnar crystals. We show that by using this method we are able to gain significant insight into the nature of high D/d packings.

Using a microfluidic flow-focusing device we generate wet foams comprised of equal volume bubbles. The bubbles in such foams may be considered spherical since they reside within the *wet region*, H_w, defined as $H_w = l_0^2/d$ where l_0 is the capillary length of the liquid and d is the bubble diameter of the foam [6].[1] Such bubbles have been previously shown to readily assume the minimum-energy configuration of hard-spheres for a wide variety of boundary conditions imposed upon them [7]. By introducing the foam into tubes of different diameters we are able to rapidly generate a variety of helical bubble assemblies. We may classify such structures by the ratio $\lambda = D/d$ where d is the equivalent sphere diameter of the bubbles and D is, again, the diameter of the tube into which the bubbles are placed.

By means of X-ray tomography we are able to access precise 3D information for individual bubbles and obtain the coordination numbers. The resulting foam packings are then compared to the hard-sphere simulations of Pickett and Mughal [2–4].

2. Experimental method

Monodisperse foam was produced using a flow-focusing device, capable of producing equal-sized bubbles of diameter between 100 μm and 800 μm through the controlled co-flow of liquid and gas [8–10]. The dispersity of bubble diameters of such samples is less than 5%, classifying these foams as monodisperse [11]. In our experiments, we used an aqueous solution composed of 5% by volume commercial detergent *Fairy Liquid* in water. We foamed using nitrogen gas into which the low-soluble compound perfluorohexane was dissolved. This significantly reduces the coarsening rate of the foam, providing the sample stability required during the imaging of the sample [12]. The bubbles container was fabricated using an *Object Eden 3D* printer composed of a polymer block (24 mm × 24 mm × 40 mm) in which 26 separate cylindrical chambers were formed. The cylinders ranged in diameter from 0.8 mm to 3.3 mm in 0.1 mm intervals. Each cylinder was 18 mm in length. This allows 26 cylindrical packings of different λ values to be imaged at the same time. By repeating the experiment several times with monodisperse foams of different bubble diameters, the variety of different λ-values imaged may be again increased.

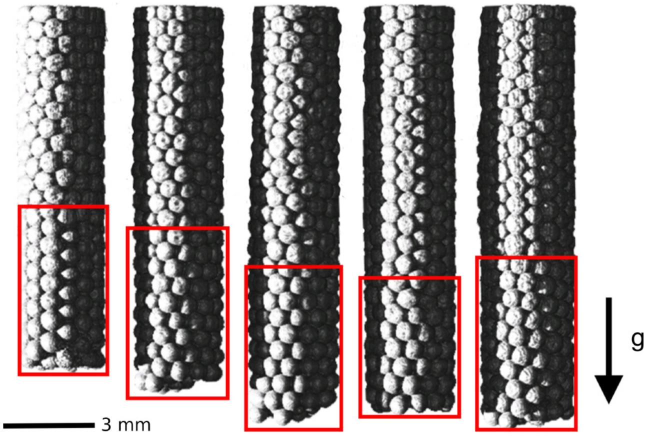

Fig. 1. Reconstruction of the raw three-dimensional tomographic data showing the cylindrical ordering of microbubbles. The boxes indicate regions of most consistent ordering. 'g' shows the direction of gravity.

The container was placed into a basin of our surfactant solution at the bottom of which was attached a flow-focusing device. The container holding the bubbles was inverted and tapped to remove trapped air, before being again inverted and placed, open face down, over the outlet of the flow-focusing device which had been previously adjusted to produce bubbles of the desired diameter. The container was closed by sliding a glass plate over the open bottom face. The resulting system was then mounted on a polyurethane plinth and allowed to rest for 2 h before being imaged. This resting period was found necessary as several bubble re-arrangements were seen to occur just after foam formation. Such movements during the image capture phase would have produced blurring in the final tomographic reconstructions.

Our tomographic device was composed of a micro-focus 150 kV Hamamatsu X-ray source with tungsten target. The sample was mounted on a precision rotation stage from Huber Germany and the sample's radioscopic projections recorded using a flat panel detector C7942 aldo from Hamamatsu(120 mm × 120 mm, 2240 × 2368 pixels, pixel size 50μm). By varying the filament voltage and current, a 100 kV filament voltage and a 100 μA were found to provide the best contrast and lowest noise in the reconstructed foam images at high spatial resolution for our experimental setup.

The tomographic images were reconstructed using the commercially available software *Octopus* [13]. The data was then analysed using the software package *MAVI* which allowed such information as bubble volume, diameter and position to be extracted [14]. Visualisation of the samples was also performed using the ray-tracing software *POV-Ray* which allows for the visual characteristics of individual bubbles to be adjusted [15]. Such software enabled the simultaneous processing and visualisation of the data; the bubbles are eventually represented by spheres of equivalent radius which may then be compared to the results of simulated annealing.

3. Results and discussion

The reconstruction of one of the samples in Fig. 1 shows foams formed in cylinders of diameter 2.9 mm, 3 mm, 3.1 mm, 3.2 mm and 3.3 mm (left to right). Each bubble on the exterior of the foam sample may be resolved. As the distance from the foam–liquid

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[1] l_0 is defined as $l_0 = \sqrt{\gamma/\rho g}$ where γ is the surface tension, ρ is the density and g is acceleration due to gravity

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