



Compressive deformation of liquid marbles

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ABSTRACT

Millimetric liquid droplets coated with particles, known as “liquid marbles”, exhibit some curious physical properties such as extended lifetime with regards to evaporation, low-friction when in contact with other surfaces and non-coalescence with other liquids. The formation dynamics have been well-characterized as well as thermal properties, however, their mechanical properties have not. Here, we investigate the response of liquid marbles to compressive deformation between two plates. Above a compressive strain of approximately 40–50%, cracks appear in the particle coating and the liquid can wet the surface of the plates, i.e. the marble ruptures. However, more strikingly, we find that even for relatively small compressive strain (without rupturing), the marbles often undergo an irreversible deformation - that is they do not regain their original shape. We quantify this shape ‘hysteresis’ across a range of particle sizes and liquids, showing that it correlates primarily with compressive strain, but also particle type and underlying fluid. Furthermore, we analyse the compressed marble shapes upon approach to rupture in the context of previous analytical approximations, showing that the recent theory of Whyman & Bormashenko (2015) provides a good description of crushed marble shapes.

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1. Introduction

Liquid marbles are millimetric droplets coated with particles [1–6]. In order to form a liquid marble, the particles must remain at the surface and most studies therefore use hydrophobic particles, although it is possible to use hydrophilic particles [7]. This shell of particles inhibits the liquid inside from making physical contact with surfaces outside, be they solid or liquid. As such, a liquid marble placed on a solid surface exhibits a non-wetting state, shown in Fig. 1. Only a slight deformation is observed at the base due to gravity [2, 3] since the marble diameter, D , is comparable to the capillary length $l_c = \sqrt{\sigma/(\rho g)}$, where σ is the liquid surface tension, ρ is the liquid density, and g is the acceleration due to gravity.

The formation of liquid marbles is an interesting phenomenon which has been studied extensively (e.g. [8–14]), eluding to some critical conditions in terms of liquid properties, and droplet to particle diameter ratio, D/d_p .

Some of the proposed applications of liquid marbles such as sensors, micro-reactors, transporters etc. [15–22], rely on the mobility of the marble, which in turn appears to depend on the packing structure of particles at the surface. If the surface coverage is too low, then wetting may occur and destroy the integrity of the marble. However, when the packing becomes too dense and all the liquid free-surface disappears, it was shown ([12–14, 23–25]) that liquid marbles undergo an irreversible deformation to a jammed interface, where non-spherical marbles

form. This is clearly an undesirable state in the context of mobility. Therefore, understanding their robustness and elasticity is paramount, yet to date, only a few studies have elucidated to mechanical properties. In particular, we note the works of [26–28] where the response to gradual compression was measured, resulting in both forces and surface areas as functions of compression ratio (versus original shape). They found that the marbles could withstand a compression of up to approximately 30%. However, in each of these studies only a single combination (liquid-particle) was studied. Bormashenko and coworkers (e.g. [29–31]) have also performed studies on the elasticity of liquid marbles, where in particular, a simple model for the shape deformation could account for the observations of elasticity in [27, 28].

Here, we report on experiments of compressive deformation of liquid marbles, similar to [27, 28], but for a wide range of liquids and particle sizes. We not only measure the critical compression ratio at which marbles rupture, but we also perform compress-release experiments and observe that even a modest compression can result in irreversible deformation. Furthermore, the simple model of [30] for the shape deformation is quantitatively tested. Whilst the rupture appears to be independent of particle characteristics and liquid, the hysteresis is found to depend on the compressive strain, particle size and underlying liquid.

2. Materials and methods

Liquid marbles are prepared by allowing a droplet of water to gently contact a bed of hydrophobic glass beads. The droplet is then manually rolled across the surface until a complete coating is achieved and subsequently placed on a horizontal microscope slide. A top plate is placed

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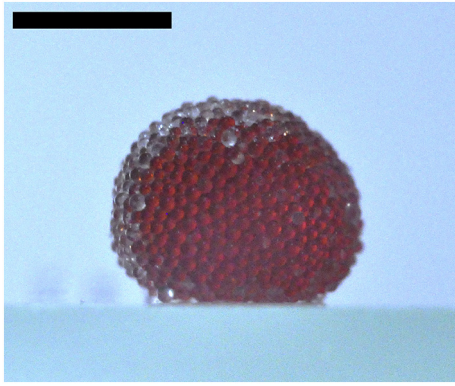


Fig. 1. A liquid marble at rest on a microscope slide. The marble is approximately 3 mm in diameter and the particles have a mean diameter of 178 μm . The scale bar is 2 mm long. Red food dye has been added for visualization purposes. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

above the marble and then gradually lowered using a motorized translation stage (MTS50-Z8, Thorlabs Inc.) at a maximum speed $dz/dt = 1 \text{ mm/s}$. The compression ratio, or vertical strain, is defined as $\epsilon_z = \Delta z/z_0$, whilst the elongation ratio, or horizontal strain, is defined as $\epsilon_x = (D - D_0)/D_0$, shown graphically in Fig. 2.

As depicted in Fig. 2, we performed two types of experiments: In the first experiment, the compression continues until the marble ruptures and the liquid wets one of the plates (Fig. 2(a)). In the second experiment, the compression was halted before rupture in order to assess the ability of the marble to regain its initial shape (Fig. 2(b)). In these compress-release cases, we characterized the deformation by comparing the initial and final shapes, i.e. $z_f - z_0$ and $D_f - D_0$. The process was captured by either high-speed video (Phantom Miro 310, Vision Research Inc.) or regular video (Nikon D90) with high-magnification to render effective pixel sizes of approximately 4.2 $\mu\text{m}/\text{px}$. The marble shapes are digitally extracted using a similar MatLab routine to that of [12], giving dimensions accurate to within $\pm 4 \text{ px}$ (2 px at each edge).

The particles comprised a range of glass beads and PTFE powders. The PTFE powder is naturally hydrophobic, whilst the glass beads were hydrophobized using the technique outlined in previous publications [12, 14, 23, 33]. The particle size distributions were characterized

Table 1

Volume-based particle size characteristics. All values stated are in μm . GB = glass beads. PTFE = Polytetrafluoroethylene powder.

Particulate label	d_{10}	d_{50}	d_{90}
GB: <53 μm	20.4	33.9	48.9
GB: 100 μm	89.2	126	169.9
GB: 100–300 μm	169	227	309
PTFE: 1 μm	0.77	1.03	1.62
PTFE: 35 μm	12.88	22.4	33.9

by the cumulative volume-based fractions (d_{10} , d_{50} , d_{90}), measured using an API Aerosizer (TSI Inc.), given in Table 1 and Fig. 3. In addition, microscope images revealed that the glass beads were circular, with $C = 4\pi A/P^2 \geq 0.9$, whilst the PTFE powders were coarse.

A water droplet placed on a layer of these powders typically exhibits an apparent contact angle between 145° and 155° , whilst images of isolated particles at the drop interface exhibit contact angles of approximately 120° (e.g. [32, 33]). NaCl and glycerin droplets exhibited slightly higher contact angles of approximately 160° and 155° respectively, however the SDS solution exhibited contact angles of approximately $87\text{--}95^\circ$ for glass beads, but $120\text{--}140^\circ$ for PTFE.

In order to study the possible influence of fluid properties, we used pure water, glycerin, surfactant and salt solutions, as detailed in Table 2. redBased upon these physical properties and radii of $R_0 \approx 1\text{--}1.6 \text{ mm}$, we estimate the following dimensionless number ranges:

$$Ca = \frac{\mu dz/dt}{\sigma} = 1.4 \times 10^{-5} \text{--} 1.2 \times 10^{-4}, \quad Bo = \frac{\rho g R_0^2}{\sigma} = 0.13 \text{--} 0.81$$

$$Oh = \frac{\mu}{\sqrt{\rho \sigma R_0}} = 0.0037 \text{--} 0.0023$$

which confirm that this process is not expected to be dominated by viscosity, but capillary forces.

In some cases, red food dye was added to the water for visualization purposes, but did not result in any measurable change in physical properties. We also note that the experiment was conducted within the timespan of one minute, redat a constant ambient temperature of 22°C so that we do not expect appreciable effects of evaporation.

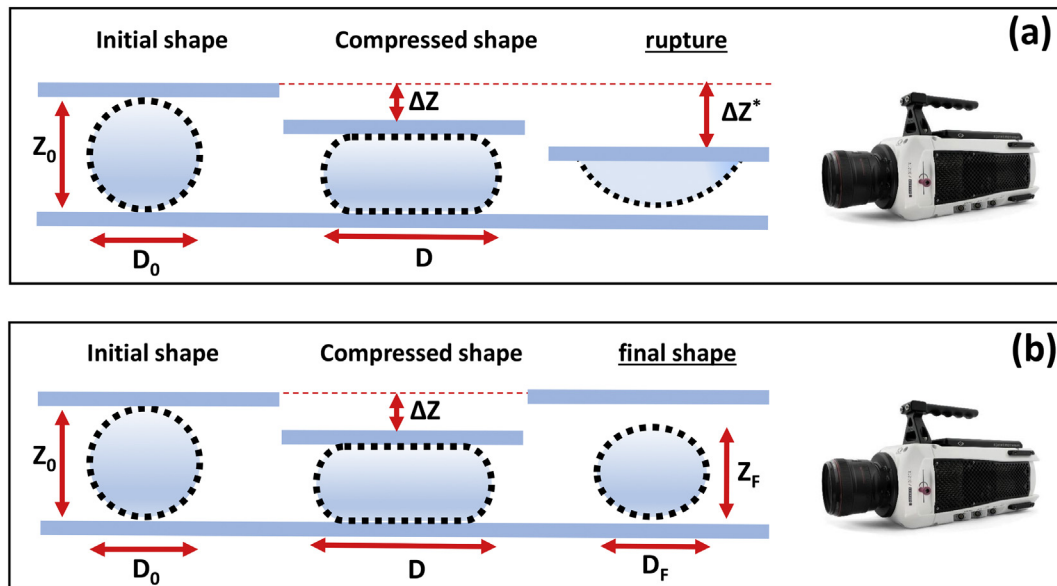


Fig. 2. Graphical definition of the measurements pertaining to the compression experiments. (a) Rupture experiment where the compressed shape upon approach to rupture is characterized by D and Δz and (b) compress-release experiment where the hysteresis is characterized by $(z_0 - z_f)/z_0$ vs. Δz .

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