Solid Drops: Large Capillary Deformations of Immersed Elastic Rods
Serge Mora, Corrado Maurini, Ty Phou, Jean-Marc Fromental, Basile Audoly, Yves Pomeau

To cite this version:

HAL Id: hal-00842852
https://hal.archives-ouvertes.fr/hal-00842852v2
Submitted on 23 Aug 2013

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L’archive ouverte pluridisciplinaire HAL, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d’enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.
Solid drops: Large capillary deformations of immersed elastic rods

Serge Mora, Corrado Maurini, Ty Phou, Jean-Marc Fromental, Basile Audoly, and Yves Pomeau

1 Laboratoire Charles Coulomb. UMR 5221, Université Montpellier 2 and CNRS. Place Eugène Bataillon. F-34095 Montpellier Cedex, France.
2 CNRS and UPMC Université Paris 06, UMR 7190, Institut Jean le Rond d’Alembert, Paris, France.
3 University of Arizona, Department of Mathematics, Tucson, USA.

(Dated: August 21, 2013)

Under the effect of surface tension a blob of liquid adopts a spherical shape when immersed in another fluid. We demonstrate experimentally that soft, centimeter-size elastic solids can exhibit a similar behavior: when immersed into a liquid, a gel having a low elastic modulus undergoes large, reversible deformations. We analyze three fundamental types of deformations of a slender elastic solid driven by surface stress, depending on the shape of its cross-section: a circular elastic cylinder shortens in the longitudinal direction and stretches transversally; the sharp edges of a square based prism get rounded off as its cross-sections tend to become circular; a slender, triangular based prism bends. These experimental results are compared to analysis and non-linear simulations of neo-Hookean solids deformed by surface tension, and are found to be in good agreement with each other.

PACS numbers: 46.25-y,68.08-p,68.35.Gy,46.15-x

We all know that liquids can be shaped by surface tension: small droplets are spherical at equilibrium so as to minimize their area, liquids climb on wet surfaces by making a smooth meniscus. The deformation of elastic solids by surface stress has been studied in more limited contexts. The macroscopic behavior of solids whose interface include features at the nanometer or micrometer scale, such as porous materials, has long been known to be influenced by the surface energy [1]. The smoothing effect of surface tension on finely textured solid interfaces has been pointed out [2, 3]. The deformations of solids near a triple line (solid-liquid-vapor) has been understood recently [4–6]. Thin structures can be deformed by capillary forces very effectively, and many examples have been studied in the past few years [7–10].

In all these examples, the effect of surface stress is amplified by the roughness of the solid or by the presence of multiple interfaces — as happens near a triple line or when an elastic structure is partially covered by a liquid drop. Inspired by the canonical examples of spherical drops and bubbles, we ask the following question: can one change the shape of a smooth elastic solid just by immersing it into a fluid? We demonstrate that this is indeed possible using a centimeter-scale piece of elastic gel. Neither surface roughness nor a triple line are required: the solid is smooth and is immersed in a uniform fluid environment. As with drops and bubbles, the deformation is driven by the change in surface tension caused by immersion. Being both soft and slender, our gels are very flexible: their deformation can be measured by simple experimental techniques and quantitatively compared to model predictions.

To deform the gel, surface tension must work against the restoring elastic forces. This competition is ruled by the elasto-capillary length \( \ell = \gamma / \mu \), where \( \gamma \) is the surface tension and \( \mu \) the shear modulus. In usual solids, both \( \gamma \) and \( \mu \) arise from phenomena at the atomic scale and \( \ell \) is of the order of a nanometer. Our gels are extremely soft, having a shear modulus as low as \( \mu = 35 \) Pa; with \( \gamma \sim 0.04 \text{ N/m} \), the length scale \( \ell \sim 1 \text{ mm} \) is large, making it possible to observe capillary effects in solids at the macroscopic scale [11–13]. The elasto-capillary length \( \ell \) is also relevant to instabilities deforming interfaces, such as the Biot [13, 14], Rayleigh-Plateau [15] and Asaro-Tiller-Grinfeld instabilities [16].

In the experiments reported below, a prismatic mould made of polystyrene is first prepared, by heating a pre-form supported by a rigid negative mould. A liquid is then introduced into the mould, which can be a hot aqueous solution of agar (from Alfa Aesar GmbH & Co) or a mixture of acrylamid and bis-acrylamid (from Merck) in aqueous solution. In both cases, a gel is formed after few hours at room temperature. The loss and storage moduli [17], measuring viscosity and elasticity, respectively, were measured in independent experiments done in similar conditions: after three hours, the ratio of the loss modulus to the storage modulus is below \( 10^{-2} \), indicating that the gel’s response has become elastic. On the time-scale of the experiment, we ignore the diffusion of the solvent inside the gel and towards the outer fluid: the gel is considered incompressible. Varying the nature of the gel and the concentration of its components, we can achieve shear moduli ranging from \( \mu = 35 \) to \( 350 \) Pa. Stress sweep tests reveal that the gel remains elastic well beyond a strain of 15\% for agar and up to 500\% for polyacrylamid gels, with a slight strain-hardening above 150\%.

A fluid interface is present along the boundary of the gel: the solvent, which remains trapped inside the gel, meets the outer fluid along this boundary. This inter-
face is associated with a surface energy ($\gamma A$), where $\gamma$ denotes the surface tension between the outer fluid and the solvent, and $A$ is the area of the boundary. The surface energy drives the deformation of immersed gels, like that of drops and bubbles. It is resisted by the elasticity of the gel, which we model as an incompressible neo-Hookean material having a density of elastic energy $w = \frac{\mu}{2} \text{tr}(\mathbf{F}^T \cdot \mathbf{F} - 1)$, where $\mathbf{F}$ is the deformation gradient, $\mathbf{I}$ the unit matrix. The equilibrium is governed by the minimization of the free energy

$$\mathcal{E} = \gamma A + \iint_{\Omega_0} \frac{\mu}{2} \text{tr}(\mathbf{F}^T \cdot \mathbf{F} - 1) \, dv_0,$$  \hspace{1cm} (1)

where $dv_0$ is a volume element in reference configuration, $\Omega_0$ is the reference volume occupied by the gel, and $A$ is the area of the deformed boundary. The incompressibility constraint writes $J = \det \mathbf{F} = 1$.

In the general theory of surface stress in solids, the surface stress is anisotropic and is a function of the surface strain [18, 19]. Our system is a special case: having a fluid origin, the capillary term in equation (1) corresponds to a surface stress that is both isotropic in the plane tangent to the boundary, and independent of the strain. The surface tension $\gamma$ between the outer fluid and the solvent can be measured independently: the comparison of experiments and models involves no adjustable parameter, even in the non-linear regime.

We start with cross-sections having the highest possible symmetry, namely circular cylinders. Cylindrical moulds with a radius ranging from $\rho_0 = 0.45$ to $2$ mm, and a length $L \approx 4$ cm are prepared, and an agar gel is formed inside the mould. Then the polystyrene mould is immersed into liquid toluene and gets dissolved within few minutes, see the inset in figure 1. The gel is denser than toluene and sinks until it reaches a horizontal grid placed at the bottom of the container. The grid is hydrophobic to prevent adhesion with the gel. The gel is imaged using a standard camera. It is always found to be shorter and thicker when immersed in toluene, compared to its initial shape set by the mould. Its surface appears to be smooth, consistent with the fact that the radius $\rho_0$ is larger than the critical radius of the Rayleigh-Taylor instability [15]. We measure the ratio $\lambda \leq 1$ of the final length to the initial length $L$, as a function of $\mu$ and $\rho_0$. The values of $\lambda$ collapse on a curve when plotted as a function of the dimensionless surface energy $\overline{\tau} = \frac{\gamma}{\rho_0 \mu} = \frac{\gamma}{\mu} \rho_0$ (figure 1). We use the independently measured water-toluene surface tension $\gamma = 36.5$ mN/m as the gel’s solvent is pure water. We have checked that the transverse expansion is consistent with our approximation of incompressibility. In view of the measured values of $\lambda \geq .85$, the agar gel remains below the elastic limit $(1 - \lambda) \sim 15\%$. The same phenomenon has been reproduced using polyacrylamid gels (data not shown).

![Image of agar gel in toluene](image)

**FIG. 1:** Shortening of an initially circular cylinder made of agar gel when immersed in toluene. The axial stretch $\lambda \leq 1$ of circular elastic cylinder is plotted, for different values for the shear modulus $\mu$ and of the initial radius $\rho_0$ (in the range $0.45$ to $2$ mm), as a function of the dimensionless surface energy $\overline{\tau}$. It is compared to the analytical formula in equation (2) (solid curve). Inset: experimental pictures for $\mu = 200$ Pa and $\rho_0 = 0.8$ mm. The agar cylinder is formed inside a translucent mould (top); after dissolution of the mould it lays on a hydrophobic grid, and shortens as the result of surface stress (bottom). To aid visualization, the boundary of the agar cylinder are highlighted using a dashed brown overlay.

The measurements of the stretch $\lambda$ can be compared to a prediction based on equation (1). Assuming that the cylinder is long ($L \gg \rho_0$) and ignoring any end effect ($L \gg \gamma/\mu$), we seek a solution in the form of a homogeneous and biaxial deformation gradient $\mathbf{F}$. We use Cartesian coordinates $(x, y, z)$, the coordinate axis $z$ being aligned with the cylinder’s axis. Owing to the cylindrical symmetry and to the incompressibility, we write $\mathbf{F} = \lambda \mathbf{e}_z \otimes \mathbf{e}_z + \lambda^{-1/2} \left( \mathbf{e}_x \otimes \mathbf{e}_x + \mathbf{e}_y \otimes \mathbf{e}_y \right)$. The deformed lateral area is $A = \left( 2\pi \rho_0^2 / \lambda^{1/2} \right) \lambda L$, where the first factor is the deformed perimeter proportional to the transverse contraction $\lambda^{-1/2}$, and the second is the length of the deformed cylinder proportional to the axial stretch $\lambda$. The area contributions coming from the disks at the ends is neglected. We insert this into equation (1), and use a thin disk $dv_0 = \pi \rho_0^2 \, dz$ as the undeformed volume element, with $0 \leq z \leq L$. The resulting expression of the energy $\mathcal{E}(\lambda) = \frac{1}{2} \pi \rho_0^2 L \mu \left( 4 \overline{\tau} \lambda^{1/2} + 2 \lambda^{-1} - 1 \right)$ is then minimized with respect to $\lambda$, which yields

$$\lambda = \left( 1 + \frac{\overline{\tau}^2}{4} \right)^{1/2} - \frac{\overline{\tau}}{2},$$

where $\overline{\tau} = \frac{\gamma}{\mu \rho_0}$. \hspace{1cm} (2)

In figure 1, this prediction is shown to yield very good agreement with the experimental data with no adjustable parameter.

We now consider prismatic rods having an initially square cross-section. After dissolution of the mould by toluene, the gel is transferred into silicon oil. Compared to toluene, silicone oils offers the advantage of having a much lower density mismatch $\Delta \rho$ with water: $\Delta \rho < 0.01$ g/cm$^{-3}$. This warrants that the ratio of the elasto-capillary length $\ell$ to the gravity length $\mu / (g \Delta \rho)$ is smaller than $5.10^{-3}$ so that gravity can be neglected. We
The rounding of an elastic wedge by surface tension has the most striking effect is that the initially square cross-section, respectively. In an isosceles but non-equilateral triangle, this point is distinct from the Spieker center of the triangle, see figure 3a. According to the theory of thin elastic rods, stretching and bending arise from the resultant force and moment of the applied load with respect to the centroid $H$ of a cross-section, respectively. In an isosceles but non-equilateral triangle, this point is distinct from the Spieker center $G$. As a result, the force equivalent to the surface tension, which is applied at $G$, induces both a compressive resultant force and a bending moment. The compressive force induces a contraction effect similar to that discussed earlier for circular cross-sections. The bending effect is novel, however, and leads to large and easily measurable displacements. Calculating the position of the points $G$
The experiment has been reproduced 3 times, yielding a non-zero bending moment. By symmetry, this bending effect disappears in the equiangular cross-sections. Dimensionless curvature \( \kappa \) of a triangular prism with apex angle \( \theta = 20^\circ \) and aspect-ratio \( L/h = 5.55 \), as a function of the dimensionless surface stress \( \bar{\gamma} \): non-linear 3d finite element simulation for a neo-Hookean incompressible material, and comparison to the linear beam theory from equation (3) for \( L \gg h \) and \( \bar{\gamma} \ll 1 \). (c') 3d shapes of the deformed prism for selected values of \( \bar{\gamma} \). The color code shows the pressure contribution \( p \) to the stress enforcing incompressibility. (c') in inset: experimental evidence of bending of agar gels immersed in silicone oil (\( h = 4.5 \text{ mm}, L = 2.5 \text{ cm}, \theta = 20^\circ, \bar{\gamma} = .0172 \)) and comparison to the corresponding finite-element simulation labeled (B).

and \( H \) and using the linear beam theory, one finds the curvature \( \kappa \) of the centerline as a function of the height \( h \) of the cross-section and of the apex angle \( \theta \) as:

\[
\kappa = \frac{\bar{\gamma}}{h} \left( \frac{1}{\sin \frac{\theta}{2}} - 2 \right) \text{ where } \bar{\gamma} = \frac{\gamma}{\mu h} . \tag{3}
\]

By symmetry, this bending effect disappears in the equilateral case (\( \kappa = 0 \) when \( \theta = \pi/3 \)), as well as for circular and square cross-sections.

This bending effect has been confirmed in experiments using agar gel immersed in silicone oil, see the inset in figure 3c’. The choice of parameters is severely constrained by the applicability of the neo-Hookean model (requiring moderate strain), the accuracy in measuring curvature (requiring large surface tension, and a good separation between the points \( H \) and \( G \)), and the need to keep the fabrication robust to imperfections: we used a length \( L = 2.5 \text{ cm} \), height \( h = 4.5 \text{ mm} \) and apex angle \( \theta = 20^\circ \). The effect of surface tension is magnified by the slenderness of the rod and is visible to the naked eye: the measured radius of curvature is \( r_{\text{exp}} = 9.5 \pm 1.5 \text{ cm} \), corresponding to a dimensionless curvature \( \bar{\pi}_{\text{exp}} = h/r_{\text{exp}} = .049 \pm .008 \). The experiment has been reproduced 3 times, yielding identical values of \( r_{\text{exp}} \) within \( \pm 0.5\% \). Given the small surface energy \( \bar{\pi} = .0172 \) (\( \gamma = 42.6 \text{ mN/m} \) and \( \mu = 550 \text{ Pa} \)) we would expect the linear theory (3) to be accurate, but it predicts a significantly larger curvature, \( \bar{\pi}_{\text{lin}} = .065 \). To explain this discrepancy, we set up FEM simulations that account both for the finite length and for non-linear elasticity, see figure 3c. The linear theory appears to be accurate in a very narrow range, \( \bar{\pi} = \ell/h \lesssim .005 \). Beyond this, the cross-sections become round and the curvature is overestimated. The non-linear simulation predicts \( \bar{\pi}_{\text{simul}} = .040 \) for \( \bar{\pi} = .0172 \), see point (B) in figure 3b, which matches the experimental value \( \bar{\pi}_{\text{exp}} = .049 \pm .008 \) up to the experimental error bounds.

Although solids are often believed to deform in a fundamentally different way to fluids, we have shown that elastic rods made of soft gels can become rounded by surface tension when immersed into a liquid, much like bubbles. This rounding has been demonstrated at the centimeter scale and involves large, easily measurable deformation. Three specific modes of deformation have been identified — they can appear concurrently, as long as the symmetry of the cross-sections allows it. A twisting mode is expected in the absence of a particular symmetry, and could be investigated in future work. The phenomenon reported here gives a way to generate large, reversible deformations simply by changing the chemical properties of a liquid bathing a solid. This effect is stronger in filamentous structures which have a large area to volume ratio and are ubiquitous in biology. This opens up the possibility of tuning the texture of a hairy interface, or actuating fibrous materials using surface tension.

---