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Depressions at the surface of an elastic spherical shell submitted to external pressure.

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Abstract: Elasticity theory calculations predict the number N of depressions that appear at the surface of a spherical thin shell submitted to an external isotropic pressure. In a model that mainly considers curvature deformations, we show that N only depends on the relative volume variation. Equilibrium configurations show single depression ($N=1$) for small volume variations, then N increases up to 6, before decreasing more abruptly due to steric constraints, down to $N=1$ again for maximal volume variations. These predictions are consistent with previously published experimental observations.

PACS: 46.70.De (Beams, plates and shells), 46.32.+x (Static buckling and instability), 89.75.Kd (Patterns).

Buckling of vessels under external pressure has been a problem addressed for a long time, as it is of utmost importance in designing tough containers in air and spatial navigation. The spherical symmetry, the simplest one that gives a close vessel, was investigated since early times [1]. Current works mainly focus on the onset of buckling and the difficulties due to non-ideality of materials [2]. For what concerns post-buckling shapes (*i.e.* adopted by the vessel when deformation keeps increasing after the critical stress), the general attention has turned to cylindrical geometry, more often encountered in applications. Recently, two papers reported about the observation of strongly buckled objects, originally porous hollow spherical shells filled with solvent, that buckle when the solvent evaporates [3, 4]. Buckling of a porous shell due to evaporation of an inner solvent is acknowledged as being of capillary origin, and macroscopically (*i.e.* at scales larger than the pore size) equivalent to the effect of an isotropic external pressure. These experiments therefore constitute a direct illustration of the postbuckling of hollow spheres under external

pressure, for which, to our knowledge, no theoretical predictions exist. Surprisingly, conformations taken by the shells qualitatively differ between the two references [3] and [4]: as shown on Fig. 1, there is occurrence of either a single and quite deep depression [3] or several depressions distributed over the sphere's surface, leading to a coarsely cubic shape [4]. The purpose of this paper is to determine whether this discrepancy could be interpreted through the equilibrium configurations of an elastic model, or if some drying artefacts should be invoked.

We will use elasticity theory results related to thin shells submitted to external constraints in order to get an insight into the number of depressions expected for an equilibrium conformation. Let us first consider one depression in a spherical elastic shell, of radius R and thickness d . With E the Young modulus of the material, the curvature constant is $\sim Ed^3$, and Ed the stretch modulus. We do not take into account energy variations linked to the gaussian curvature, as its integral on a closed surface depends only on the topology

(Gauss-Bonnet theorem). It has been shown [5, 6] that a depression corresponds to the inversion of a spherical cap, which avoids stretching energy (preponderant for shells of nonzero thickness) out of the circular ridge that joins the undeformed part and the inverted cap, defined by its half-angle α (Fig. 2). Minimization of the elastic energy concentrated in the ridge imposes its lateral extension $\delta \sim (Rd)^{1/2}$. Pauchard et al showed that the relevant curvature radius in this region is $\delta/\tan \alpha$ [7]. The energy of a single depression therefore writes:

$$U_1 = 2 \pi E d^3 (d/R)^{-1/2} \sin \alpha \operatorname{tg}^2 \alpha \quad (1)$$

This expression diverges when α approaches $\pi/2$: then, stretching deformations are likely to release the high energy cost of a ridge with infinite curvature. However, as will appear obvious later, other considerations different from energetic ones prevail in this limit, and looking for a more accurate expression is unnecessary within the current work.

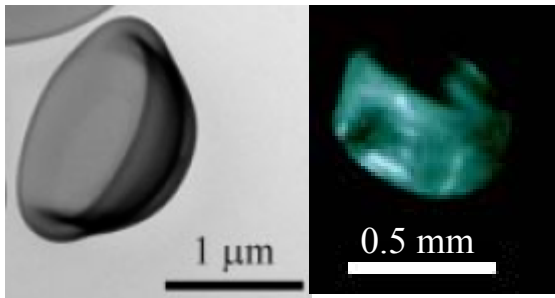


Figure 1: Different shapes obtained after evaporation of the solvent contained in a spherical porous shell. Left: silica/silicon « capsule » ($N=1$) observed by Zoldesi et al [3] (reproduced with author and editor permission). Right: more polyhedral shape ($N \sim 6$) observed by Tsapis et al in shells made from aggregation of colloidal particles at the surface of an evaporating droplet of colloidal suspension [4] (reproduced with author permission).

The volume variation ΔV due to cap inversion is twice the volume of the spherical cap, hence the volume variation relative to the undeformed sphere volume:

$$(\Delta V/V_{\text{sph}})_{1 \text{ depression}} = (1 - \cos \alpha)^2 (2 + \cos \alpha)/2 \quad (2)$$

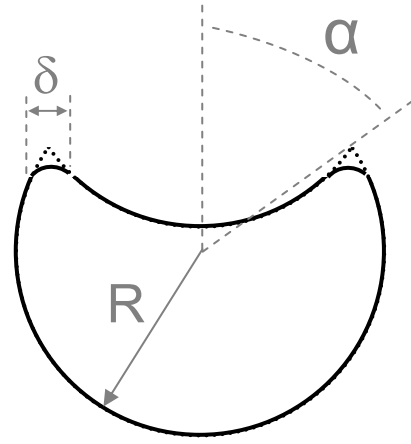


Figure 2: Depression formed by inversion of a spherical cap. The circular ridge that allow a continuous jonction between the undeformed spherical part and the inverted cap has a lateral extension $\delta \sim (Rd)^{1/2}$ (where d is the thickness of the shell). The aperture of the depression is defined by the half-angle α that extrapolates (dotted line) the ridge thickness down to zero.

In the case of N similar depressions, we have:

$$U_N = N U_1 \quad (3a)$$

and

$$\Delta V/V_{\text{sphere}} = N (\Delta V/V_{\text{sphere}})_{1 \text{ depression}} \quad (3b)$$

We therefore have the expressions of both the elastic energy and the volume variation corresponding to N similar depressions formed by inversion of spherical caps of half-angle α . As the relative volume variation is the key parameter to appreciate the deformation intensity, it would be interesting to eliminate α in order to get the elastic energy U_N as a fonction of $\Delta V/V_{\text{sphere}}$, and then to discuss the relative stability of the conformations with different numbers N of depressions (“states”).

Explicit expression for small depressions:

For small inverted caps ($\alpha \ll 1$), system (3) simplifies in:

$$U_N \sim 2 \pi N E d^{5/2} R^{1/2} \alpha^3$$

and

$$\Delta V/V_{\text{sphere}} = N \alpha^4 / 8$$

Which leads, for a given relative volume variation $\Delta V/V_{\text{sphere}}$:

$$U_N \sim 8^{13/12} \pi N^{1/4} E d^{5/2} R^{1/2} (\Delta V/V_{\text{sphere}})^{3/4} \quad (4)$$

For a given shell (determined E , d and R), the $N^{1/4}$ dependence with $\Delta V/V_{\text{sphere}}$ clearly leads

$N=1$ at equilibrium. The conformation with a single depression is favored in this regime,

which corresponds to small deformations ($\Delta V/V_{\text{sphere}} \ll 1$).

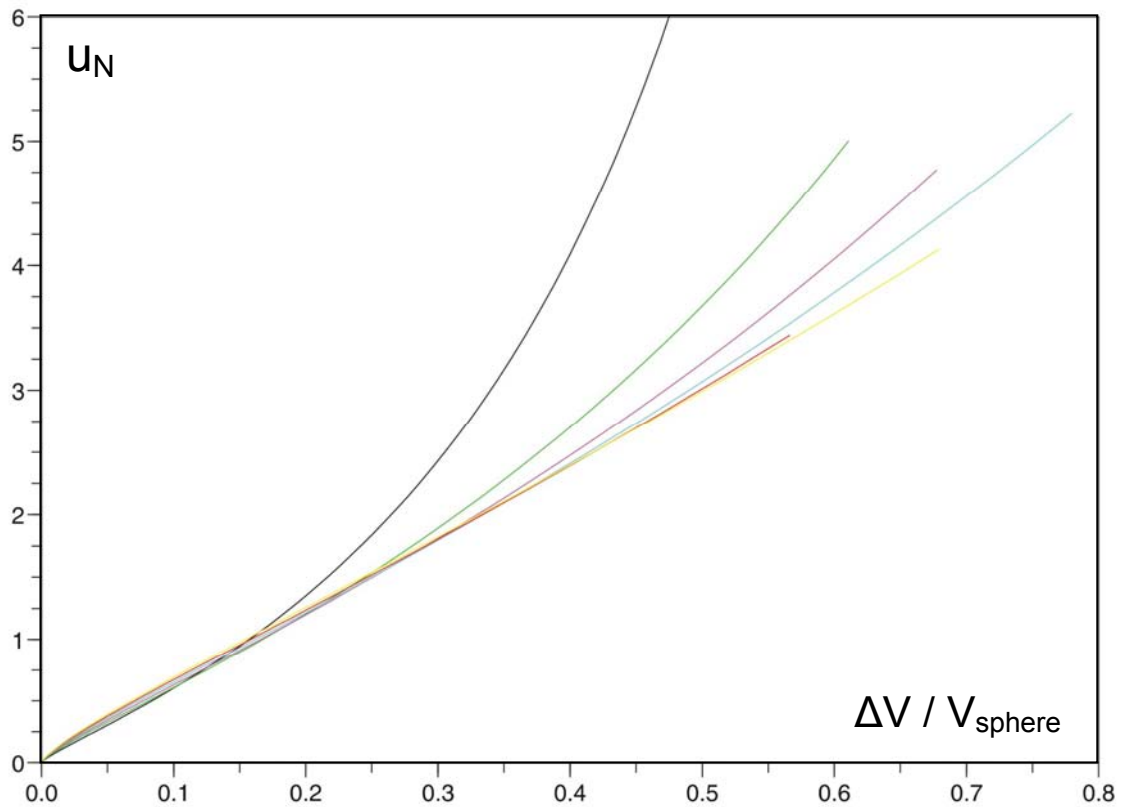


Figure 3: Reduced energy $u_N = U_N / (2 \pi E h^3 (d/R)^{-1/2})$ as a function of $\Delta V/V_{\text{sphere}}$, for different numbers N of similar depressions (inverted spherical caps). The curves are traced up to the maximum value $(\Delta V/V_{\text{sphere}})_{\text{max},N}$ over which there would be an interpenetration between depressions. Black, green, pink, blue, red and yellow curves correspond respectively to $N = 1$ to 6. One recognizes the $(\Delta V/V_{\text{sphere}})^{3/4}$ behaviour of eq. (4) for small $\Delta V/V_{\text{sphere}}$, that correspond to small values of α .

Implicit dependence for larger depressions:
For larger values of α , a parametric plot of U_N as a function of $\Delta V/V_{\text{sphere}}$ shows that $N=1$ is

no longer the lowest energy state when $\Delta V/V_{\text{sphere}}$ increases, but $N=2$, then $N=3$ etc (Fig. 3). However, system (3) is not sufficient

to describe the conformations adopted at high deformations, since equation (3b) is valid only if depressions do not interpenetrate each other. There is of course a maximum size (*i.e.* maximum α) over which it is not possible to find an arrangement of N similar depressions that avoids interpenetration between two of them. For the sake of simplicity, we will neglect the thickness of the circular ridge, which corresponds to the case $d \ll R$. Then the problem is a pure matter of geometry: how many similar spherical caps can be inverted without interpenetration at the surface of a sphere?

A necessary condition is that the spherical caps themselves do not overlap before inversion. This amounts to search which maximum surface of a sphere can be covered with N similar spherical caps, which is another formulation of the so-called Tammes' problem: maximize the minimum point-to-point distance for a set of N points placed on a sphere [8]. The resolution of the Tammes' problem is much less obvious than its formulation; analytical or numerical solutions exist [9-12], that for our purpose allowed to calculate $(\Delta V/V_{\text{sphere}})_{\text{max}}$ for $N \geq 5$ (Table 1).

For up to $N = 4$, the necessary condition is not sufficient. This is obvious for $N = 2$: the caps corresponding to the solutions of the Tammes' problem are two hemispheres ($\alpha = 90^\circ$), which cannot be reverted both simultaneously. Interpenetration of two opposite depressions begins when they contact at the center of the sphere, which corresponds to a maximum value of the depression half-angle α : $\alpha_{\text{max},2} = 60^\circ$. We will make the same sort of operation in what follows: keeping the centers of the spherical caps of the Tammes' problem solutions at their positions, then decrease α up to a value for which there is not interpenetration. For $N=3$, the caps that solves the Tammes' problem are centered on the vertices of an equilateral equatorial triangle (this can be easily understood, as the intersection between the plane defined by the three points and the sphere is a circle. Three points on a circle maximize their mutual separation with an equilateral spreading, and the point-to-point distance increases with the circle size, which is maximum when the circle is equatorial); three lines of trigonometric

considerations show that interpenetration is avoided when α reduces to $\alpha_{\text{max},3}$ such as: $\cos \alpha_{\text{max},3} = 1/\sqrt{3}$ (Fig. 4). In a similar way, one can show for $N=4$ that $\cos \alpha_{\text{max},4} = 1 / (2 \sin \alpha_T/2)$, where $\alpha_T = 109^\circ$ is the angle between two center/vertex directions in the tetrahedron. Hence the half-angle defining a spherical cap reduces from 54.59° (Tammes' problem solution for $N=4$, with tetraedric symmetry) to 52.24° for the non-interpenetrating inverted caps. For higher values of N ($N > 4$), the polyhedron holding the bases of the spherical caps present only right or obtuse angles between adjacent faces: there is therefore no risk of interpenetration as long as the caps bases do not overlap – and the Tammes' condition is sufficient.

With these values of $\alpha_{\text{max},N}$, the maximum relative volume variation $(\Delta V/V_{\text{sphere}})_{\text{max}}$ could be calculated for each N , allowing the energy U_N of the state with N depressions to be plotted only for relative volume variations that have, for each N , a physical meaning. This is shown on Fig. 3, for $N=1$ to 6. Higher values of N are not represented, since they all correspond to energies superior to U_6 for physical values of $\Delta V/V_{\text{sphere}}$ (*N.B.*: for N higher than the values displayed on Table I, the asymptotic formula for upper limit $\alpha_{\text{max},N}$ (rad) = $(2\pi/\sqrt{3})^{1/2} N^{-1/2}$ [13] shows that it keeps decreasing).

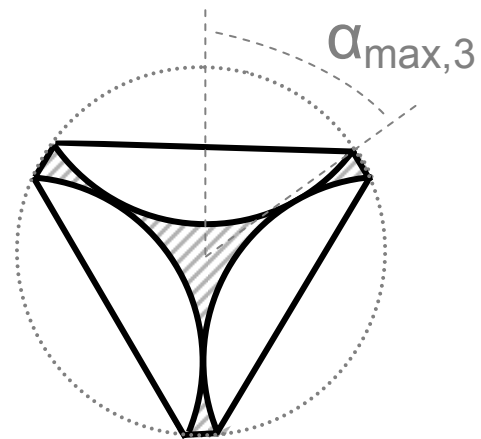


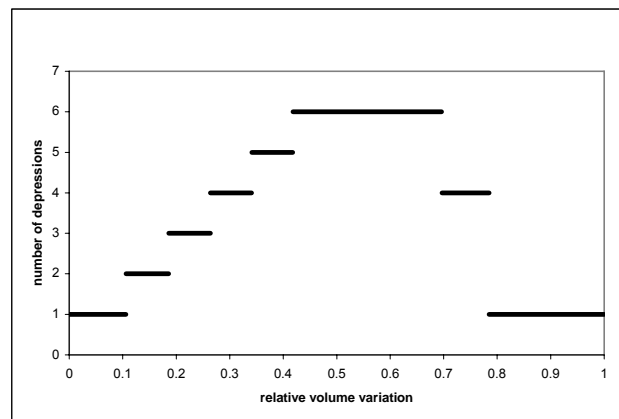
Figure 4: Equatorial section of the $N = 3$ conformation: $\alpha_{\text{max},3} = 54,74^\circ$. The solution to Tammes' problem would correspond to slightly larger spherical caps ($\alpha = 60^\circ$) that could not revert without interpenetration.

N	α_{\max} (deg)	maximum $\Delta V/V_{\text{sphere}}$
1	90	1
2	60	0.625
3	54.7	0.691
4	52.2	0.785
5	45	0.581
6	45	0.697
7	38.9	0.480
8	37.4	0.474
9	35.3	0.427
10	33.1	0.373
11	31.7	0.350
12	31.7	0.382
13	28.6	0.277
14	27.8	0.270
15	26.8	0.251

Table I: Maximum values $\alpha_{N,\max}$ of the half-angle α that defines a spherical cap at the surface of a sphere, over which one cannot invert N similar caps without interpenetration. Values of α_{\max} for cases $N=1$ to 4 are treated in the text. Values for $N=5$ to 8 are given in reference [10], $N=9$ in reference [12], $N=10$ to 15 in reference [11]. As explained in the text, higher values of N will appeared as useless for our purpose. Third column: the corresponding volume variation for the N inverted caps.

Looking for the lowest energy at every relative volume variation provides the phase diagram displayed on Fig. 5: in the early steps, the number of depressions increases with $\Delta V/V_{\text{sphere}}$ due to energetic considerations, from $N=1$ up to a quite stable cubic organization of the depressions ($N=6$). Such an evolution clearly is in accordance with observations by Gao *et al* [14] on polyelectrolytes capsules submitted to an increasing osmotic pressure. Then steric factors favorizes a tetraedric-related conformation ($N=4$), followed by the ultimate single depression state. It is interesting to note

that the state $N=1$ for high relative volume



variations is made compulsory by the geometry: this explains *a posteriori* why there is no point in refining the energy calculation in this limit.

Figure 5: Equilibrium values of the number of similar depressions N at the surface of the sphere, as a function of the relative volume variation $\Delta V/V_{\text{sphere}}$. The five first transitions occur at $\Delta V/V_{\text{sphere}} = 0.106, 0.185, 0.274, 0.341$ and 0.418 . The following “steric” transitions occur to values given in Table I for $N=6$ and $N=4$.

In the model derived up to now, a single parameter (the total relative volume variation) happens to drive the conformations; this is worthwhile to be commented. As previously stated, we did not consider the extension of the circular ridge that continuously links the inverted cap to the spherical undeformed part. Taking it into account would (i) increase for each depression the volume variation by a term scaling in $\delta^2 R$, which means a correction in $N(d/R)$ for $\Delta V/V_{\text{sphere}}$ (ii) lower the maximum surface compactness of depressions, by adding an excluded surface around the caps, which amounts to decrease $d_{\max,N}$ and then $(\Delta V/V_{\text{sphere}})_{\max,N}$ for each N . If the prefactor of d/R in the volume correction varies slowly with α , the global effect of (i) will be a displacement of the transitions towards smaller values of $\Delta V/V_{\text{sphere}}$, proportionally to d/R . By affecting $(\Delta V/V_{\text{sphere}})_{\max,N}$, (ii) will cause an additional displacement of the “steric” transitions (*i.e.* transitions towards smaller values of N). Stretching energy has also been neglected by hypothesis. This could begin to be discussed for the most acute bendings that

occur at high relative volume variations, but we saw that in this limit the phase diagram is driven by steric, rather than energetic, considerations.

Another point of importance is that the present model is purely static. Like in numerous buckling problems, the system can be stucked in metastable states, especially here for steric transitions that need a high energy barrier merging of distinct depressions. This is likely to explain why the final $N=1$ state could be reached only for a few capsules in reference [14].

In conclusion, we showed that a quite simple model of curvature deformations at equilibrium is sufficient to make the first predictions concerning postbuckling conformations of a spherical shell submitted to an external pressure. In this model, the parameter that drives the transitions between different states (*i.e.* different number of depressions formed by inversion of a spherical cap) is the total relative volume variation. The buckled sphere exhibits a single depression for both small and important relative volume variations, and several depressions (up to 6, which leads to a cubic symmetry) for intermediate ones. These results are consistent with experimental observations: “capsules” with a nearly hemispheric single depression for important variations of the inner volume, observed by Zoldesi et al, or coarsely cubic shapes for weaker volume variations, observed by Tsapis et al. To make more accurate predictions, a more sophisticated model considering both the geometry of the ridge and stretching deformations is required. It should reveal, at higher orders, an influence of the relative shell thickness d/R on the boundaries of the phase diagram.

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