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EPL, 106 (2014) 24004

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Image: Ornamental multiplication of space-time figures of temperature transformation rules (adapted from T. S. Bíró and P. Ván 2010 EPL 89 30001; artistic impression by Frédérique Swist).
Secondary polygonal instability of buckled spherical shells

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received 5 March 2014; accepted in final form 7 April 2014
published online 22 April 2014

PACS 46.32.+x – Static buckling and instability
PACS 46.70.De – Application of continuum mechanics to structures: Beams, plates and shells
PACS 46.25.-y – Static elasticity

Abstract – When a spherical elastic capsule is deflated, it first buckles axisymmetrically and subsequently loses its axisymmetry in a secondary instability, where the dimple acquires a polygonal shape. We explain this secondary polygonal buckling in terms of wrinkles developing at the inner side of the dimple edge in response to compressive hoop stress. Analyzing the axisymmetric buckled shape, we find a compressive hoop stress with parabolic stress profile at the dimple edge. We further show that there exists a critical value for this hoop stress, where it becomes favorable for the membrane to buckle out of its axisymmetric shape, thus releasing the compression. The instability mechanism is analogous to the formation of wrinkles under compressive stress. A simplified stability analysis allows us to quantify the critical stress for secondary buckling. Applying this secondary buckling criterion to the axisymmetric shapes, we can determine the critical volume for secondary buckling. Our analytical result is in close agreement with existing numerical data.

Introduction. – All spherical elastic shells, such as sports and toy balls or microcapsules, exhibit a qualitatively identical deformation behaviour upon deflation: At small deflation, the capsule remains spherical. Below a critical volume, the classical buckling instability occurs, and an axisymmetric dimple forms [1–3]. Finally, this dimple loses its axisymmetry in a secondary instability, resulting in a polygonal buckled shape (see fig. 1). This deformation behaviour is seen on the macroscale for elastomer balls, on the microscale in experiments on microcapsules [4–8] and for pollen grains [9], as well as in computer simulations based on triangulated surfaces [6,10,11] or finite element methods [12–14]. The same sequence of an axisymmetric buckling instability followed by a secondary polygonal buckling instability also occurs when a dimple is formed by indenting the capsule with a point force [13–15], when the capsule is pressed between rigid plates [14] or when the capsule adheres to a substrate [16]. Even droplets with a colloidal shell, which are initially fluid and not elastic, show similar buckling phenomena [17–19].

The first buckling transition, where an axisymmetric dimple forms, is well understood. Linear shell theory can be successfully used to calculate the onset of instability of the spherical shape [2,3]. Furthermore, nonlinear shell theory has been used to investigate the post-buckling behaviour, which revealed that the buckled shape is unstable with respect to further volume reduction if the pressure is controlled [2,20,21]. Numerical analyses of nonlinear shape equations have been used to characterise the bifurcation behaviour of axisymmetric shapes [21,22]. The results of ref. [21] show that the first buckling transition exhibits a bifurcation behaviour that is analogous to a first-order phase transition, with the volume for the onset of instability (the spinodal) differing from the critical volume where the elastic energy branches of spherical and buckled shapes cross.

In contrast, the theory of the secondary buckling transition, where the dimple loses its axisymmetry, has remained mostly phenomenological based on the existing results from computer experiments [6,11]. A theory rationalizing the underlying mechanism and predicting the critical volume of secondary buckling is still lacking. In this letter, we show that secondary buckling is caused by compressive hoop stresses that occur in the
inner neighbourhood of the dimple edge after axisymmetric buckling. In order to release the compressive stress, the circumferential fibres buckle out of their circular shape if the hoop stress reaches a critical value; this instability is similar to wrinkling under compressive stress [1] or comparable to the Euler buckling of straight rods [2]. The quantitative investigation of the secondary buckling transition therefore consists of two steps: Firstly, determining the stress distribution in the axisymmetric buckled configuration, and secondly, finding the critical compressive stress at which the axisymmetric configuration loses its stability.

The first task can be accomplished by numerical integration of the shape equations derived from nonlinear shell theory [21] or by an analytic approach based on the ideas of ref. [23]. The second task necessitates an analysis of the stability equations of shallow shells [3]. In this letter, we focus on the mechanism of secondary buckling and the resulting parameter dependences of the critical buckling stress. Together with geometrical relations, these nonlinear differential equations — called shape equations — can be solved numerically [21].

For the analysis, it is convenient to introduce dimensionless quantities by using $EH_0$ as the unit for tensions and $R_0$ as the unit length. Specifically, this results in a dimensionless bending stiffness $E_B \equiv E_B/EH_0R_0^2 = H_0^2/R_0^2(1 - \nu^2)$, which is the inverse of the Föppl-von-Kármán number $\gamma_{FVK} = 1/E_B$.

For a qualitative understanding of the shape and stress distribution of an axisymmetric buckled capsule we start with vanishing bending stiffness $E_B = 0$. Then, the equilibrium shape consists of a mirror inverted spherical cap (see fig. 2(c), gray lines), which is isometric to the initial spherical shape and, therefore, unstrained. For $E_B > 0$, the sharp edges of the inverted cap give rise to an infinitely large bending energy. Hence, these sharp edges must be smoothed out.

Upon smoothing, see fig. 2(c), the inner neighborhood of the edge is shifted to the inside, towards the axis of symmetry, and the outer neighborhood is shifted to the outside. Circumferential material fibers will be compressed in the inner neighborhood and stretched in the outer neighborhood; but far away from the dimple edge, we expect the deformation to decay. This draws a qualitative picture of the circumferential stress distribution $\tau_\varphi(s_0)$ along the arc length: It has a zero at $s_D$ (the arc length position of the edge, see fig. 2), a positive maximum for $s_0 > s_D$ and a negative minimum for $s_0 < s_D$; it approaches zero for $s_0 \to 0$ and $s_0 \to \infty$ (cf. fig. 3).

Along these lines, Pogorelov constructed an analytic model for axisymmetric buckled shapes [23]. To describe this deformation from the isometric shape to the final smooth shape, he introduced a displacement $(u(s_0), v(s_0))$ in $r$- and $z$-direction, respectively, see fig. 2(c). Assuming $u$ and $v$ to be small, linear shell theory can be employed to calculate the bending and stretching energies in the final shape by means of calculus of variations with respect to $\delta u$ and $\delta v$ (with some simplifications). From the approximate solutions $u(s_0)$ and $v(s_0)$ presented in [23], analytical expressions for curvatures, tensions and stresses

![Fig. 2: Geometry of the axisymmetric midsurface. (a) Un-deformed shape (always with index “0”); (b) deformed shape; (c) Pogorelov model: close-up of the smooth final shape (black) with small displacements $(u, v)$ to the isometric shape (grey).]

In these equations, $q$ is the transversal shear force, and $p$ the applied normal pressure, which can also be interpreted as a Lagrange multiplier to control the capsule volume. Together with geometrical relations, these nonlinear differential equations — called shape equations — can be solved numerically [21].

The deformed shape is given by

$$0 = \frac{\cos \psi}{r} \tau_\varphi + \frac{1}{r} \frac{d(r\tau_\varphi)}{ds} - \kappa_\varphi q,$$

$$0 = -p + \kappa_\varphi \tau_\varphi + \kappa_\varphi \tau_\varphi + \frac{1}{r} \frac{d(rq)}{ds},$$

$$0 = \frac{\cos \psi}{r} m_\varphi - \frac{1}{r} \frac{d(rm_\varphi)}{ds} - q.$$
1. Which is close to the minimum of the exact numerical equations. The total elastic energy is found to be of the final shape can be deduced, which are generally

2. Simplication of the compressed strip as a plate.

3. Of the final shape can be deduced, which are generally

4. Simplification of the compressed strip as a plate. (c) Simplication of the compressed strip as a plate.

5. Of the final shape can be deduced, which are generally

6. Simplification of the compressed strip as a plate.
is the critical stress at which wrinkling occurs,
\[
\tau_c = a_p E_B \hat{\tau}_c (\hat{a}_c) \quad \text{with} \quad \hat{\tau}_c = \sqrt{EH_0/E_B a_c a_p^{-3/2}}, \quad (7)
\]
where the function \( \hat{\tau}_c (\hat{a}_c) \) is known numerically, see fig. 4.

Our analysis also shows that the secondary buckling transition is a \textit{continuous} transition in the sense that the wrinkle amplitude \( W \) at the transition can remain arbitrarily small [24]. This is in contrast to the primary buckling transition, which is a \textit{discontinuous} transition with metastability above and below the transition [21] and with an axisymmetric dimple of the buckled state which always has a finite size.

**Phase diagram for deflated spherical capsules.** –

The function \( \hat{\tau}_c (\hat{a}_c) \) generated this way can now be applied to the stability analysis of the axisymmetric buckled capsule shapes. For a given numerical solution of the axisymmetric shape equations, we have to compute the parameters \( a_p \) and \( a_c \), calculate the critical buckling stress according to (7) and compare it to the minimum value \( \tau_{\text{min}} = \min_{a_p} \tau_c (s_0) \) of the hoop stress in the compressive region. If \( \tau_{\text{min}} < -\tau_c \), then the capsule cannot bear the compression and will form polygonal wrinkles, losing its axisymmetry.

The curvature parameter \( a_c \) is, by definition, \( a_c = \kappa_c (s_c) \) where \( s_c \) is the root of \( \kappa_c \). As mentioned beforehand, the parameter \( a_p \) for the parabola of the stress profile is to be determined by the condition that the approximating parabola has the same integral over the compressive region as the original stress function \( \tau_c (s_0) \). Let \( F = \int_{s_1}^{s_2} \tau_c (s_0) ds_0 \) denote this integral, which has the physical interpretation of the net force in the compressive region \( s_0 \in [s_1, s_2] \). It can be evaluated numerically for a given solution. For a parabola of the form \( \tau_{\text{par}} = -\tau_c (1 - a_p (s_0 - s_c)^2) \), one finds \( a_p = (4\tau_0 /3F)^2 \), which is to be inserted into (7).

In our numerical analysis, we applied this scheme to axisymmetric buckled shapes with different bending stiffnesses \( E_B \) and reduced volumes \( \Delta V/V_0 \). We control the volume rather than the pressure, since for given pressure the capsule buckles through [21]. In this case, the secondary buckling might take place in a modified form. For each value of \( \bar{E}_B \), the critical capsule volume, where the criterion \( \tau_{\text{min}} < -\tau_c \) for polygonal buckling is fulfilled, is determined numerically. This critical volume for the secondary buckling transition is shown in the phase diagram, fig. 5 (red dots). Fitting the data points with a power law \( i.e. \) a straight line in the double logarithmic phase diagram yields

\[
(\Delta V_{2nd}/V_0)_{\text{shape exp.}} = (2550 \pm 50) \bar{E}_B^{0.946 \pm 0.002} \quad (8)
\]

with an exponent close to \(-1\).

Analysing the Pogorelov model with our secondary buckling criterion, we can also derive a simple analytical expression for the critical volume where the secondary buckling occurs. We find the following analytical results
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Fig. 5: (Colour on-line) Phase diagram of deflated spherical capsules with Poisson ratio \( \nu = 1/3 \) in the plane of reduced bending stiffness \( \tilde{E}_B \) and reduced volumes \( \Delta V/V_0 \) (double logarithmic). Dots represent results from the shape equations. The blue and red lines represent the critical volumes of first and secondary buckling, respectively, according to the Pogorelov model. The green line is the classical result for axisymmetric buckling. Dashed lines are results for the secondary buckling according to computer simulations (refs. [6,11]).

for the three parameters of the plate buckling criterion,

\[
\tau_0 \sim \frac{EH_0}{1-\nu^2} \left[ \tilde{E}_B (1-\nu^2) \frac{\Delta V}{V_0} \right]^{1/4},
\]

\[
a_p \sim R_0^{-2} \left[ \tilde{E}_B (1-\nu^2) \right]^{-1/2}, \quad a_c \sim \left[ 1 - \nu^2 \frac{\Delta V}{V_0} \right]^{1/4} \tilde{E}_B E_0 \frac{\sqrt{1-\nu^2}}{2}. \]

Using these scaling results in the secondary buckling criterion (7) (treating \( \tau_c (\tilde{a}_c) \) as a numerical factor) yields \( \Delta V/V_0 \sim \tilde{E}_B B \). A detailed calculation which includes the prefactors yields

\[
(\Delta V_{2nd}/V_0)_{\text{Pog}} = 3706 \tilde{E}_B, \tag{9}
\]

where the exponent 1 is exact [24]; the prefactor still weakly depends on \( \nu \) and is given here for \( \nu = 1/3 \). This result is very close to the results from the shape equations (see fig. 5, red line).

For both models, the number of wrinkles can be obtained by comparing the critical wavelength \( \lambda_c \) (fig. 4) to the perimeter \( 2\pi r(s_c) \) of the parallel on which the wrinkles form. In the Pogorelov model we obtain 5 wrinkles, independent of the bending stiffness; using the shape equations, we find between 8 (for small \( \tilde{E}_B \)) and 6 wrinkles (for larger \( \tilde{E}_B \)). In experiments on deflated microcapsules, a wider range of wrinkling numbers can be observed, from three (or even two, corresponding to an elongated dimple) up to at least eight [4–6]; however, these shapes were not observed at the onset of the secondary buckling but showed well-developed wrinkles. Numerical simulations in ref. [6] produced three to six wrinkles at the onset of buckling; with three and four wrinkles appearing only in simulations of the re-inflation of a wrinkled shape. A more detailed discussion is postponed to ref. [24].

Figure 5 also shows, in dashed lines, results of computer simulations for the critical volume of the secondary buckling, which can be fitted by power laws \( \Delta V/V_0 = 3400 \tilde{E}_B^0 [6] \) and \( \Delta V/V_0 = 8470 \tilde{E}_B^{0.85} [11] \) with exponents close to 1. Results from our secondary buckling criterion match the simulation results fairly well, although the parabolic stress profile is only a rough approximation of the real circumferential stress. In ref. [24] we will present a numerical stability analysis of the full axisymmetric shape, without the parabolic approximation, which confirms the present results.

The phase diagram is supplemented by corresponding lines for the first buckling transition from a spherical to an axisymmetrically buckled shape. The classical buckling line (green line in fig. 5) is derived from the well-known classical buckling pressure \( p_{cb} = -4\sqrt{Eh_0 E_B}/R_0^2 \) [1–3]. It describes the pressure at which the spherical configuration becomes unstable and is equivalent to a volume difference

\[
\Delta V_{cb}/V_0 \approx 6(1-\nu)\tilde{E}_B^{1/2} \tag{10}
\]

with an exponent 1/2 [11]. This line coincides very well with the data points from the shape equations (green points in fig. 5), which were taken at the volume where the axisymmetric buckled shapes branch off the spherical shapes [21].

The axisymmetric buckled state is unstable if pressure is controlled instead of volume [2,20,21], because the load that the capsule can bear is getting smaller when the dimple grows. Thus, for given pressure, the dimple that forms at the classical buckling pressure \( \tilde{E}_{cb} \) grows spontaneously until a shape with stable pressure-volume relation is found. For all bending rigidities considered in ref. [21], this only happens if the dimple gets in contact with the opposite side of the capsule.

Already for volume differences \( \Delta V/V_0 \) smaller than that of the classical buckling transition (10), the spherical shape is only metastable. From the solutions of the shape equations, we can compute the smallest volume difference where the branch of axisymmetric buckled shapes becomes energetically favourable to the spherical solutions (blue points in fig. 5) [21]. For small \( \tilde{E}_B \), this critical volume difference is substantially smaller than that of the classical buckling transition, thus leaving a large volume region (between the two lines) where the spherical shape is metastable and the axisymmetric dimpled shape is the global energy minimum. Koiter’s stability analysis [20] suggests that the buckling transition of real (imperfect) shells occurs somewhere in this region, depending on the severity of imperfections.

Pogorelov’s model can also be used to calculate the volume where the elastic energies of the dimpled shape (5) and spherical shape (see ref. [6]) are equal, which gives

\[
\frac{\Delta V_{1st}}{V_0} \bigg|_{\text{Pog}} = 6J^{4/5}(1-\nu)^{4/5}(1-\nu^2)^{-1/5} \tilde{E}_B^{3/5} \tag{11}
\]

for the critical volume of the first buckling transition with an exponent 3/5. This result is in close agreement
with the data points from the shape equations (see fig. 5, blue line).

Conclusions. – In this letter, we explained the mechanism underlying the secondary buckling instability of an initially spherical elastic capsule including a quantitatively correct value for the critical capsule volume. This completes our theoretical understanding of the generic deformation behaviour of spherical capsules upon volume reduction, which starts with a spherical shape for small volume changes, then jumps to an axisymmetric buckled shape in a primary buckling transition, and finally results in a non-axisymmetric shape with polygonal wrinkles along the inner neighbourhood of the dimple edge after the secondary buckling transition.

So far, the secondary buckling transition has only been observed in experiments or simulations but was lacking a physical explanation. The key ingredient underlying the secondary buckling is a locally compressive hoop stress, with a characteristic negative peak near the edge of the axisymmetric dimple. We conducted a quantitative analysis, in that we approximated the profile of the compressive hoop tension $\tau_1$ by a parabola. This led to a derivation of a critical compressive stress, quite analogous to the critical value for the secondary buckling is proportional to $\Delta S_{2}$. These results are in good agreement with all existing numerical simulation data except numerical results in ref. [10], where $\Delta S_{2} \sim \frac{V}{V_{0}}$ is found. This differing result might be caused by using a vanishing equilibrium curvature $\kappa_{s0} = \kappa_{\phi0} = 0$ in the elastic energy of the simulation model in ref. [10].

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