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Shaping gels and gels mixture to create helices

Special Collection: Kitchen Flows

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ABSTRACT

In cooking, food gels, such as agar-agar or alginate, are often prepared and presented in the form of spheres or spaghetti. While experimenting in our kitchen, we realized that it is quite difficult to make more advanced shapes. In this study, we sought to develop new methods to obtain more complex shapes. Our first challenge was to obtain helices. The best method we selected was to deposit the solutions before their gelation in a thread. The robustness of the method is tested by systematically changing the thread pitch, diameter, and depth. From the deformation under its own weight, we propose to deduce the mechanical characteristics of the helix. These values are compared to those obtained in the laboratory using indentation testing. Finally, we experimented with mixed gels obtained by combining agar-agar and alginate.

I. INTRODUCTION

When the first lockdown started, we wanted to take this opportunity to finally get into molecular cooking, which mixes culinary art and science. A direction that is currently still under-researched in the field of molecular cooking concerns the influence of food shapes on the eating experience. This issue is central to the food industry and research in this area has now established that shapes influence our perception of food.[1–3] A limiting point is the reduced number of shapes that can be produced by the techniques used in molecular cooking. In this article, we show that it is possible to obtain complex shapes from the ingredients already used in molecular cooking and with low-cost materials and techniques.

Molecular cooking is a recent branch of culinary arts that uses scientific approaches from physics, chemistry, or biology to obtain novel flavors, textures, and new foods. Several starred chefs are exploring this new trend, and new recipes have already been created. One of the directions being explored is to use the acquired knowledge on the physics and chemistry of gelation—a general way of converting a fluid into a solid—to produce new shapes like spheres and noodles, and to encapsulate flavors.[4] Gelation is an important process not only in the food industry, but also in a wide range of other applications, such as waste treatment,[5] agriculture,[6] and environmental engineering,[7,8] and has potential application in medicine[9,10] and material sciences.[11] Gels usually contain many solvent molecules like water (50% to 90%) trapped inside a network, which confers to it mechanical properties identical to those of an elastic solid. The network is made up of molecules, polymers, particles, colloids, etc., that are cross-linked at specific parts on them.[12] Recent studies have shown that mechanical properties can be improved by mixing two gels.[11–13] This technique requires the use of components with different gel conditions allowing the creation of a first network in which the second one is entangled. This method is not yet used in the kitchen or in the food industry, and we will see that it can be employed to do a two-step sequential shaping.

In cooking, the most popular methods used to obtain gels are by using sodium alginate or agar-agar. Sodium alginate, the sodium salt of alginic acid, is soluble in water and forms hydrogels in the presence of divalent cations, such as Ca^{2+}, through cation binding.[14] There are two methods to create spheres depending on the calcium content of the product to be encapsulated in the gel.[15] When the substance contains no calcium, sodium alginate is added and the substance is "cooked" in a bath of calcium. The reverse (we talk then of external spherification) is preferred when the substance contains calcium ions. In this case, the substance is cooked in a sodium alginate bath and the gelation reaction stops when the spheres are removed. Spheres are also obtained with agar-agar, a product extracted from certain species of red seaweeds[16,17] that has practically no taste nor color. Agar consists of a mixture of two polysaccharides: agarose (∼70%) and agaropectin (∼30%). Agarose, which is at the origin of the gel formation, is a polymer made up of repeating units of agarobiose. The molecule appears to have a double helical structure; furthermore, the gelling property of

agarose is due to hydrogen atoms, which constrain the molecule to form an helix.[11,13] When the agar-agar solution is heated to 90 °C and left to cool down, helices are formed with a subsequent association and reorganization of the helices, which creates a three-dimensional network via helical bundles at a temperature of about 40 °C. This phase transition is thermoreversible. To make spheres, drops of hot agar-agar solution are formed and left in a "cold" not water-miscible liquid, typically oil, forming gel spheres. Straight fibers or spaghetti can be obtained by allowing the agar to cool in a tube. In this study, we investigate whether it is possible to realize more complex shapes, such as a helical filament using the gels and the tools present in our kitchens.

One way to create helical shapes is by deforming flat surfaces. This is done by using layers of hydrogels with different swelling properties[18] or by positioning high-swelling gels in a non-swelling gel[19,20] that deform when exposed to stimuli. Helical shapes are also obtained by ionoprinting of thermosensitive hydrogels that create periodic undulations in the mechanical properties.[21] Gradient of cross-linking agents density[22] or the use of different cross-linking agents is also possible methods.[23,24] In the above-mentioned cases, the design requires advanced manufacturing techniques, light-activated reactions, or 3D/4D printing. Those methods are not yet available in the kitchen. At our disposal, we have a set of pasta paddles used to roll pasta. One of the paddles has a groove that looks like a screw channel. It appears that placing a strand of alginate solution in the groove prior to immersing in a calcium bath is a very promising solution. After a few hours, it is quite easy to peel off the gel. Based on this first observation, we present two new methods to generate flexible helices made of sodium alginate or a mixture of alginate with agar-agar. These methods consist in casting an alginate solution (eventually added with agar) in a helical mold prior to immersing it in a bath of calcium lactate. Section II details the methods to produce helices. Then, we present the helices resulting from these methods along with a characterization of their mechanical properties. In particular, we measure the deformation of the helices when hung vertically, which allows us to deduce their Young's modulus. Finally, we discuss how these results compare with previous measurements obtained with the same hydrogels and give some perspectives to these methods.

II. EXPERIMENTAL METHODS

A. Mold fabrication

To reproduce and optimize the groove geometry, we 3D printed molds with helicoidals grooves (see Fig. 1) using a polylactide material. The typical geometry of the molds is shown in Fig. 1. We use molds with different geometric parameters, which are detailed in Table I. The molds allow us to vary the helix diameter D, pitch p, and length ℓ_0, and we keep constant the groove size.

In a second step, the device is improved by adding a removable sheath that can be placed around the first mold. The sheaths are also 3D printed and have a slightly larger diameter than the diameter D of the mold. This technique allows the use of the most liquid gels.

B. Protocol for the gels

In this work, we focus on two different types of gels: pure sodium alginate and a mixture of sodium alginate with agar-agar. As we have seen in the introduction, sodium alginate can form a gel either by immersing the solution in a calcium ion bath or by immersing a fluid rich in calcium ion in an alginate bath. Both methods are tested, but we will present experiments performed with the first method (dipping sodium alginate in a calcium ion bath). In Secs. II B 1 and II B 2 describes the preparation of the different solutions and the method followed to obtain the helices.

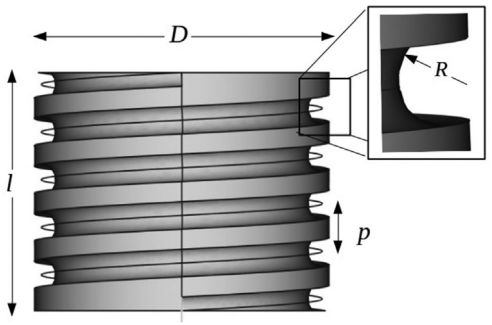

FIG. 1. Sketch of the molds. They are first drawn with FreeCAd and then printed with a 3D printer. D: diameter of the mold, ℓ_0: height of the mold, $d = 2R$: groove diameter, and p: pitch.

1. Method used to obtain sodium alginate helices

We prepare sodium alginate solutions from food grade products obtained from QUIMICA M.G. The sodium alginate powder is first dissolved in water (pH 6) at ambient temperature ($T = 20$ °C) until all lumps disappear. We use three concentrations: 20, 30, and 40 g/l. Drops of food coloring are added to the solution to identify the sodium alginate concentration. Above a concentration of 40 g/l, the alginate solution is very viscous and it is difficult to remove all lumps and to obtain an homogeneous preparation. For this reason, we do not study solutions with concentrations above 40 g/l.

For concentrations of 30 and 40 g/l, the solution is sufficiently viscous so that the groove can be filled without the liquid leaking out. A syringe is used to place the solution into the groove, and once filled, the mold is immersed for 5 min into the calcium lactate bath (20 g/l) at ambient temperature. We verify that leaving the alginate longer in

TABLE I. Geometric parameters of the molds used to make the helices. ℓ_0: length, D: diameter, d: diameter of the groove, and p: pitch.

Mold	ℓ_0 (mm)	D (mm)	d (mm)	p (mm)
1 H20-6	24	20	5	6
2 H15-7	30	15	4	7
3 H10-6	24	10	4	6
4 H11-7	30	11	4	7
5 H8-7	30	8	4	7
6 H10-5	30	10	4	5
7 H8-5	30	8	4	5
8 H15-5	30	15	4	5
9 H10-10	30	10	4	10
10 H8-10	30	8	4	10
11 H15-10	30	15	4	10

the calcium bath does not significantly affect the mechanical properties of the gel. The mold is removed from the bath and washed with clean water. To prevent the drying of the gel, the mold is then placed in an air-tight box for 10 h protected from changes in temperature or humidity. This duration is chosen to ensure that the gelling process has occurred throughout the whole section of the helix.

For the 20 g/l sodium alginate solution, the process should be slightly modified, as the solution flows very easily and drains out of the mold before gelation begins. In this case, the molds are immersed in the alginate solution and placed in the freezer, in such a way as to allow the solution to begin to solidify, but not completely. Once the desired solidification point is reached, the excess sodium alginate is removed from the outside of the mold with the help of a spatula and immediately immersed in the calcium lactate bath. The concentration of 20 g/l is the minimum concentration for which it is still possible for the alginate to gel in the mold before flowing out.

2. Helices obtained by mixing agar-agar and sodium alginate

Two protocols are used to obtain a mixture of agar-agar and alginate gels. The first protocol consists in heating the alginate solution up to a temperature of 90 °C before adding the agar powder and stirring the preparation until complete dissolution. Then, the solution is left cooled down and the agar-agar gel forms with alginate molecules embedded inside the network. For the second protocol, the agar solution is first heated to 90 °C before incorporating the alginate solution prepared at room temperature. The solutions are prepared with 20 g/l of alginate mixed with 10 or 20 g/l of agar-agar. No significant difference between the gels is noticed.

We carried out rheological measurements (rotational rheometer Anton Paar MCR 501 with a cone-plane geometry) to confirm that the sodium alginate molecules are not affected by the heat. Figure 2 shows the results for a solution of 20 g/l of sodium alginate, which is heated for 10 min at different temperatures and then cooled again.

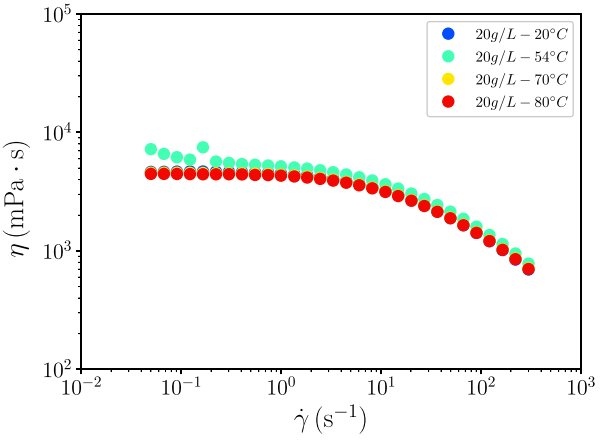

FIG. 2. Dynamic viscosity η as a function of the shear rate $\dot{\gamma}$ of 20 g/l alginate solutions that are heated to different temperatures during 10 min before left cooled down. The measurements are made after the solution had cooled at room temperature $T = 20$ °C.

We see that the rheological property is not impacted by the different heating conditions. The alginate solution shows, whatever the treatment, a shear-thinning behavior for shear rates above 10 s^{-1}.

When the mixture is then immersed in a calcium bath, we observed that the alginate forms a second network that allows the gel to take a new shape. We use this property to create helices. For this, the agar-agar/alginate mixture while its cooling ($T \gtrsim 50$ °C) is placed in a syringe. The syringe is connected to a plastic tube into which the solution was injected. The tube is then placed under cold water or left at room temperature for the solution to cool and the gel to form. This procedure usually takes a few minutes. Then, air is injected into the tube with an empty syringe, which allows the gel to come out. The gel retains the shape of the tube. The filament is then placed inside the groove of the mold. After that, the mold is immersed in the calcium lactate solution.

3. Measurements of the elastic properties using indentation testing

The mechanical response of the sample is characterized using nanoindentation testing (Anton-Paar). The method consists in driving a spherical tip of radius $R_i = 200$ μm into the sample with a controlled penetration depth, δ. The spherical tip, assumed to be undeformable, is initially in contact with the surface of the gel. Then, the tip is driven inside the sample with a loading speed of 10 μm/min until a maximum load is reached. The applied force, F, is measured as a function of the penetration depth, δ. Measurements are reported in Fig. 3 for several samples. The resulting loading curves are well fitted by the Hertz contact law revealing the elastic behavior of the material:

$$F = \frac{4E\sqrt{R_i}}{3(1-\nu^2)}\delta^{3/2}, \tag{1}$$

where E is the Young's modulus and ν is the Poisson's ratio of the sample ($\nu = 0.4$ for alginate gels[25]). The fit of the indentation-force curve with a Hertz law provides an estimate of the Young's modulus of alginate gels.

III. EXPERIMENTAL RESULTS

Figure 4 presents three helices made with the same mold and pure alginate gels of increasing concentrations. We note that the alginate gel adopts a well-defined helical shape whatever the alginate concentration. A helix obtained with a mixture of alginate and agar-agar gels is shown in Fig. 5. Here again, the gel filament is observed to be a well-shaped helix. One of the main differences between the two methods is that the second produces filaments of perfectly circular cross sections whereas the first method leads to cross sections that are closer to a semicircle.

The helices are flexible and may experience large deformations when handled vertically. By assuming that they deform like elastic springs, we will show that the Young's modulus of the gel can be estimated from the measurement of this deformation. This simple test will enable us to compare the mechanical properties of different gels. We will first describe the procedure and the result for alginate gels. Then, the method will be extended to gel obtained by mixing agar-agar and alginate.

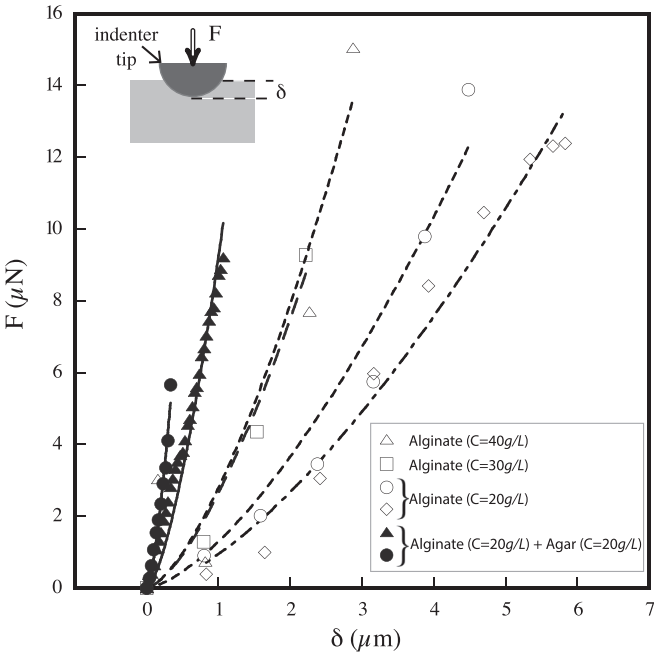

FIG. 3. Applied load, F, as a function of the penetration depth, δ, of the indenter tip on different gels. The Young modulus of the gels are obtained by fitting the data with the Hertz law [see Eq. (1)]. A symbol represents a sample. Solid and dashed lines are adjustment of the data by Eq. (1).

A. Measurement of ℓ_h

In order to measure the length ℓ_h of the helical filament in the absence of stress, the helices were left to sediment slowly in a container filled with water. In this way, contact forces with a solid substrate or stresses due to gravity are reduced and ℓ_h can be estimated with a better precision. The corresponding mold is then placed in the container next to the gel helix. An image with both the mold and the filament is then taken. The image is opened using the ImageJ.[40] A line parallel to the axis of the mold is drawn by hand, and the two ends are carefully placed to match the ends of the mold. The software gives the length ℓ_0 of the mold in pixel. The same operation is repeated for the helix, which gives us the length of the helix ℓ_h. The length of the mold is used to calibrate the lengths. Figure 6 shows the picture of one helix made of alginate along with its mold. We see that the helix matches quite well the groove of the mold: The pitch, the length, and the diameter are very close. Figure 7 displays ℓ_h normalized by the length ℓ_0 of the mold used to fabricate the helix. In the case of pure alginate [Fig. 7(a)], the rest length of the helix is slightly larger than the mold length whatever the concentration in alginate. Figure 7(b) shows the same measurements for helices made of alginate ($c = 20$ g/l) and agar-agar gels with two concentrations. Despite the fact that results are more

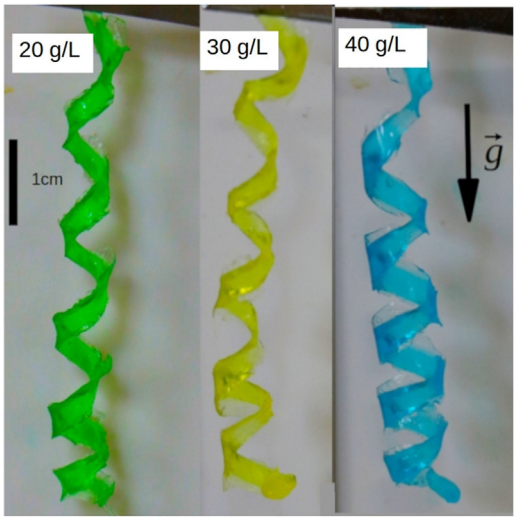

FIG. 4. Three alginate helices suspended under their own weight. The alginate concentration is, respectively, from left to right 20, 30, 40 g/l. The length of the vertical bar is 1 cm.

FIG. 5. Helix made of alginate (20 g/l) and agar-agar (10 g/l) suspended under its own weight. The white bar represents 5 mm.

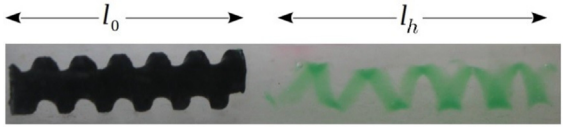

FIG. 6. Helix made of alginate gel ($c = 20$ g/l) immersed in water. The black mold used to shape the helical filament stands at the left. The mold length is $\ell_0 = 30$ mm and corresponds to mold number 5 in Table I.

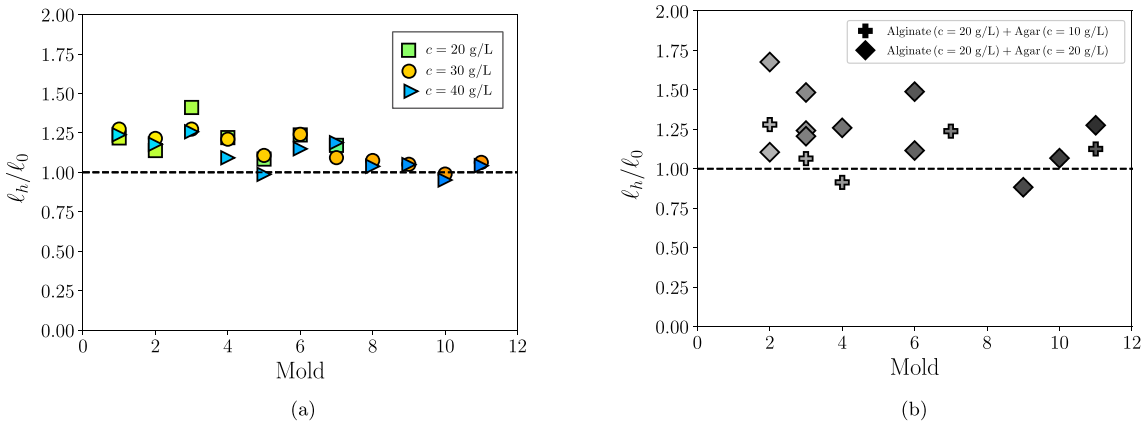

FIG. 7. (a) Estimated length ℓ_h of pure alginate helices after sedimentation in a water normalized by the length ℓ_0 of the mold. Green squares, yellow circles, and blue triangles correspond to helices obtained with, respectively, a concentration of 20, 30, and 40 g/l in alginate. The indices of the x-axis refer to mold reference given in Table I. (b) Same results for helices obtained with a mixture of alginate at 20 g/l and agar-agar gels. Crosses and diamonds represent measurements made for agar-agar concentrations of 10 and 20 g/l, respectively.

dispersed with gel mixtures, the ratio ℓ_h/ℓ_0 is also generally larger than one in this case.

B. Measurement of ℓ

To measure ℓ, the helices are suspended by one end and a ruler is placed close to the helices. Images are then taken and opened with the ImageJ.[40] In the same way as for determining ℓ_h or ℓ_0, a segment is drawn along the fiber axis with its ends as close as possible to the fiber ends. The pixel length of the segment ℓ is then converted into millimeters using the ruler present in the image. Figure 8 shows the vertical deformation $(\ell - \ell_0)/\ell_0$ of hanging helices made of pure alginate (30 g/l) as a function of the diameter D and for different pitches p. In this case, additional molds are used, whose geometric parameters are observed in Fig. 8. Note that the length of the helix clamped is subtracted from ℓ. We observe that for a given concentration in alginate and a given diameter, the normalized deformation of the helix increases with its diameter and reduces with its pitch. Thus, diameter and pitch appear to be critical parameters for the mechanical properties of these flexible helices.

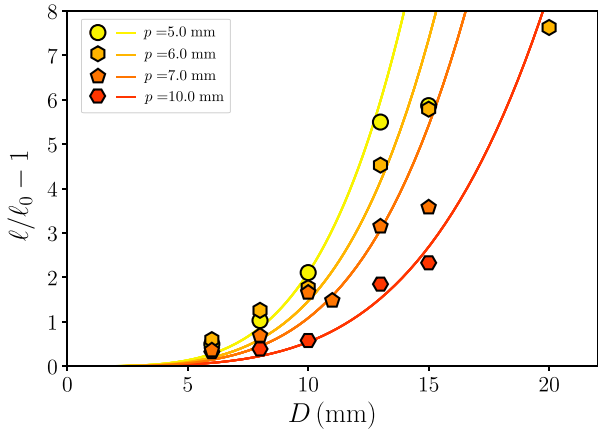

FIG. 8. Normalized vertical deformation of the helix hanging on its own weight as a function of the helix diameter D for different pitches p, $d = 4$ mm. The helices are made with a concentration $c = 30$ g/l in alginate. Solid lines correspond to the predictions of Eq. (4) for the different pitches and $E = 72$ kPa.

C. Model

In order to rationalize our observations, we develop a mechanical description of the helix deformation under its own weight. The helix is assumed to be made of a filament with a cylindrical cross section (diameter d), an elastic material of density ρ, and Young's modulus E. At rest, the diameter, pitch, and length of the helix are, respectively D, p, and ℓ_0. The stiffness k of the helix is given by the relation[26]

$$k = \frac{Ed^4 p}{8D^3 \ell_0}, \tag{2}$$

and the total mass M of the helical filament writes

$$M = \rho \frac{\pi d^2 \ell_0}{4} \sqrt{1 + \left(\frac{\pi D}{p}\right)^2}. \tag{3}$$

When the helix is submitted to its own weight, it extends by a length $\Delta\ell = \ell - \ell_0$, which expresses $\Delta\ell = \frac{Mg}{2k}$. Combining these equations, we get a prediction for the normalized vertical length $\ell/\ell_0 = (\ell_0 + \Delta\ell)/\ell_0$,

$$\frac{\ell}{\ell_0} = 1 + \frac{\pi \rho g \ell_0 D^3 \sqrt{1 + \left(\frac{\pi D}{p}\right)^2}}{Ed^2 p}. \tag{4}$$

In order to compare this prediction with our measurements, we can plot the vertical deformation $(\ell - \ell_0)/\ell_0$ as a function of the quantity $\mathcal{P} = \pi \rho g \ell_0 D^3 \sqrt{1 + (\frac{\pi D}{p})^2}/d^2 p$. A linear variation of the data will indicate an elastic regime. We can then get the elastic modulus as the inverse of the slope of the data. If the cross section of the filament

is semi-circular (like for alginate helices) rather than circular, the elastic modulus can be estimated by plotting the vertical deformation as a function of $\tilde{\mathcal{P}} = 2\mathcal{P}$. This is because the decrease in the cross section reduces the mass M by a factor 2 and the stiffness k by a factor 4, resulting in a decrease in the normalized deformation by a factor 2.

D. Estimations of Young's modulus

We first present the estimation of the Young's modulus deduced from the vertical deformation. Figure 9(a) shows the vertical deformation for alginate helices as a function of the parameter $\tilde{\mathcal{P}}$. We observe that the vertical deformation increases with $\tilde{\mathcal{P}}$. When $\tilde{\mathcal{P}} \lesssim 200$ kPa, the increase is linear and it becomes sub-linear for larger values of $\tilde{\mathcal{P}}$. The solid line shows the linear fit of the experimental data for $\tilde{\mathcal{P}} < 200$ kPa. The inverse of the slope provides an estimation for the Young's modulus. For alginate, we found $E = 72 \pm 8$ kPa. The experimental data do not reveal any clear dependence of E with the alginate concentration. The prediction of the model for $E = 72$ kPa is plotted in Fig. 8. We observe a good agreement between the experimental data and the model, except for the experiments correspond to high values of $\tilde{\mathcal{P}}$. In this case, the deformations are lower than the theoretical predictions.

Results for helices obtained by mixing alginate and agar-agar are shown in Fig. 9(b). The dispersion of these results reflects the greater difficulty to fabricate these helices. The wrapping of the soft and fragile filament into the helical mold is a difficult step. Even so, we observe a linear variation of the helices deformation as a function of $\mathcal{P}$ and a weak effect of the agar-agar concentration on the variation of the deformation with $\mathcal{P}$. We also observe that the elastic limit is reached for $\mathcal{P} \gtrsim 700$ kPa a much higher value than for alginate. The best linear fit of these data gives an estimate of the Young's modulus for the alginate and agar-agar mixtures of $E = 270 \pm 20$ kPa. The double gelation process produces stiffer helices and increases the range in which the linear response is observed.

Indentation tests were performed on fragments of the helices: some made of pure alginate and some obtained by mixing alginate and agar-agar. The fit of the indentation-force curve with a Hertz's law (see Fig. 3) provides an estimate of the Young's modulus. For the alginate we found: $E = 92 \pm 40$ kPa, with no clear dependence of E with the alginate concentration. The dispersion around the mean value is mainly due to differences in E from one sample to another. We also note that indentation test gives a 20% higher value compared to the Young modulus estimated from the vertical deformation (92 kPa instead of 72 kPa). For mixed gels, the indentation test gives: $E = 873 \pm 301$ kPa. This estimate is consistent with the fact that gels obtained by mixing alginate and agar-agar give stiffer helices. However, this value is three times higher than the one obtained from the deformation measurement. As for the measurements on alginate, the values obtained with the indenter are very dispersed. (The dispersion is always about 40% of the average.) We believe that this is due to the fact that the indenter is used in the limit of its range of use.

IV. DISCUSSION

The mechanical properties of alginate hydrogels determined can be compared with previous measurements. Compression experiments realized on alginate beads prepared in the same range of concentration provide a Young's modulus between 5 and 300 kPa, in relative agreement with our observations.[27,28] However, these experiments show a slight increase in E with the alginate and the cation concentrations in contrast with our experiments where the impact of the gel concentration is not significant. Other studies show that the mechanical properties of alginate hydrogels also depend on the ionic strength of the solutions and the storage environment.[29,30] Thus, it would be interesting to explore in further details how these parameters impact the mechanical characteristics of our helices.

Another property of alginate gels that has been characterized in the literature is the limit of linear elasticity above which the material becomes strain-hardening.[28,31,32] This limit is reached for a typical strain $\varepsilon \sim 0.4$. For the soft helices, the global deformation $(\ell - \ell_0)/\ell_0$ is related to the maximal local deformation ε by the relation $\varepsilon \propto d/\ell_0\,(\ell/\ell_0 - 1)$ as the filament is solicited in bending. In this study, the ratio ℓ_0/d is roughly constant and equals to 8 ± 1; thus, the limit of elastic deformation is reached for helix deformations $(\ell - \ell_0)/\ell_0 \sim 3$. This is in reasonable agreement with the observations shown in Fig. 9

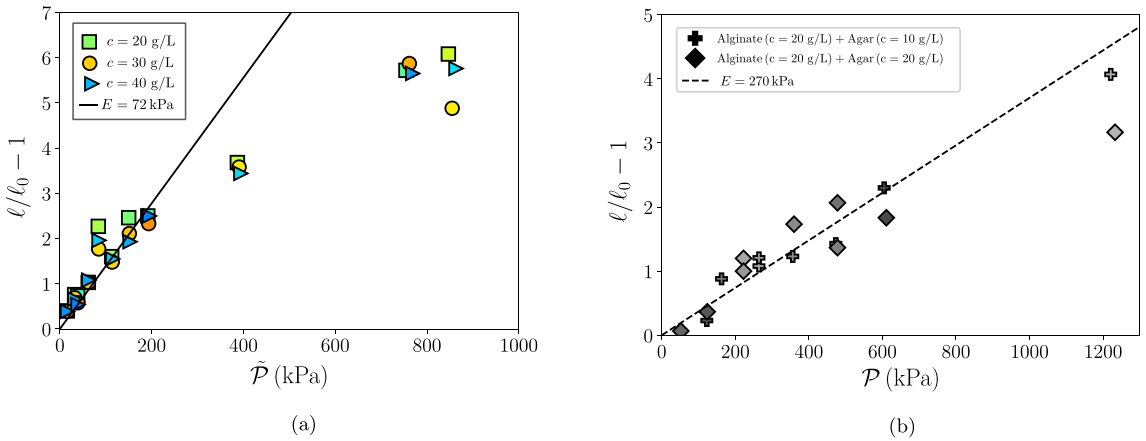

FIG. 9. (a) Normalized vertical deformation of alginate helices $(\ell - \ell_0)/\ell_0$ as a function of the parameter $\tilde{\mathcal{P}} = 2\pi\rho g \ell_0 D^3 \sqrt{1 + \left(\frac{\pi D}{p}\right)^2}/d^2 p$ for different concentrations c in alginate. (b) Same results for helices made of alginate ($c = 20$ g/l) mixed with agar-agar ($c = 10$ g/l: crosses and $c = 20$ g/l: diamonds) as a function of $\mathcal{P}$. Symbols and colors are identical to those in Fig. 7.

for alginate and mixed gels, which both deviate from a linear trend when $(\ell - \ell_0)/\ell_0 \gtrsim 3$.

The second method allows to obtain helices with a regular section and a very clean shape. The difficulty that we have encountered with this method concerns the wrapping phase around the mold. In order to not break the alginate/agar filament, it is necessary that this operation be done slowly and carefully, especially for the smaller diameter molds.

As noted in the introduction, it is also possible to create a gel by dipping a solution containing calcium ions in an alginate bath. We then talk of external spherification. We also test the possibility of using this method to stiffen agar-agar and make it take a new shape. First, calcium lactate (30 g/l) is dissolved in a warm liquid solution containing 10 g/l of agar-agar. We then use the same method as before. After cooling, the agar-agar is rolled onto a mold and then dipped into solution containing 5 g/l of sodium alginate (instead of a calcium lactate bath as previously). With these conditions, we do not obtain a gel strong enough to keep the desired shape. However, we believe that this approach is promising but requires further study to optimize the different concentrations to be used.

V. CONCLUSION AND PERSPECTIVES

In this study, we propose two "recipes" allowing us to shape filaments made of hydrogels into helices, the first with pure alginate and the second with a combination of alginate and agar-agar gels, both available in most kitchens. These methods enable the creation of helices with a wide range of length, pitch, and diameter and different gel concentrations. We also propose a simple test to characterize the mechanical properties of these helices: measuring their vertical deformations when they hang under their own weight. Assuming that the helices behave as elastic springs, we estimate the Young's modulus of these objects and we find estimates in reasonable agreement with independent indentation tests as well as the estimates given in the literature. Finally, this work proves that it is possible to impose a natural curvature to gels, which have very low rigidity. This opens new avenues to morph food[33–36] and to fabricate complex shapes with soft gels, a fact which might be of interest for soft robotics, bio-encapsulation, and 3D printing techniques.

Indeed, soft robotics are now looking for new manufacturing methods in order to shape biocompatible and biodegradable materials and reduce their environmental footprint.[37] On the other end, alginate encapsulation of molecules of biological significance, such as enzymes, drugs, macromolecule, micro-organism, or stem cells, is a well-established method.[38,39] Our study shows that more complex shapes can be obtained from the mixture of two different gels; this could have applications in this field by increasing for example the surface to volume ratio and increasing the exchanges between the capsule and the environment.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Maria Veronica D'Angelo: Conceptualization (equal); Writing – review and editing (equal). **Ludovic Pauchard:** Writing – review and editing (equal). **Harold Auradou:** Writing – review and editing (equal). **Baptiste Darbois Texier:** Writing – original draft (lead).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

REFERENCES

[1]A. Normann, M. Röding, and K. Wendin, "Sustainable fruit consumption: The influence of color, shape and damage on consumer sensory perception and liking of different apples," Sustainability **11**, 4626 (2019).
[2]S. C. Scheele, C. Hartmann, M. Siegrist, M. Binks, and P. F. Egan, "Consumer assessment of 3D-printed food shape, taste, and fidelity using chocolate and marzipan materials," 3D Print. Addit. Manuf. (published online, 2021).
[3]S. Zhou, S. Chen, and S. Li, "The shape effect: Round shapes increase consumers' preference for hedonic foods," Psychol. Mark. **38**, 2051–2072 (2021).
[4]*Handbook of Molecular Gastronomy*, edited by R. M. Burke, A. L. Kelly, C. Lavelle, and H. T. vo Kientza (CRC Press, 2021).
[5]B. G. A. Mueller and A. Sarkar, *New Membranes and Advanced Materials for Wastewater Treatment* (American Chemical Society, 2009).
[6]A. E. Al-Rawajfeh, M. R. Alrbaihat, and E. M. AlShamaileh, "Characteristics and types of slow- and controlled-release fertilizers," in *Controlled Release Fertilizers for Sustainable Agriculture*, edited by F. Lewu, T. Volova, S. Thomas, and R. K. R. (Academic Press, 2021), Chap. 4, pp. 57–78.
[7]S. Kazakov, M. Kaholek, I. Gazaryan, B. Krasnikov, K. Miller, and K. Levon, "Ion concentration of external solution as a characteristic of micro- and nano-gel ionic reservoirs," J. Phys. Chem. B **110**, 15107–15116 (2006).
[8]B. P. Spalding, S. C. Brooks, and D. B. Watson, "Hydrogel-encapsulated soil: A tool to measure contaminant attenuation in situ," Environ. Sci. Technol. **44**, 3047–3051 (2010).
[9]P. Li, Y. F. Poon, W. Li, H.-Y. Zhu, S. H. Yeap, Y. Cao, X. Qi, C. Zhou, M. Lamrani, R. W. Beuerman, E.-T. Kang, Y. Mu, C. M. Li, M. W. Chang, S. S. J. Leong, and M. B. Chan-Park, "A polycationic antimicrobial and biocompatible hydrogel with microbe membrane suctioning ability," Nat. Mater. **10**, 149–156 (2011).
[10]A. Dodero, S. Alberti, G. Gaggero, M. Ferretti, R. Botter, S. Vicini, and M. Castellano, "An up-to-date review on alginate nanoparticles and nanofibers for biomedical and pharmaceutical applications," Adv. Mater. Interfaces **8**, 2100809 (2021).
[11]Q. Chen *et al.*, "A robust, one-pot synthesis of highly mechanical and recoverable double network hydrogels using thermoreversible sol-gel polysaccharide," Adv. Mater. **25**, 4174 (2013).
[12]H. Henning Winter, *Gel Point* (Wiley, 2016), pp. 1–15.
[13]Q. Chen, "Fundamentals of double network hydrogels," J. Mater. Chem. B **3**, 3654 (2015).
[14]T. Funami, Y. Fang, S. Noda, S. Ishihara, M. Nakauma, K. I. Draget, K. Nishinari, and G. O. Phillips, "Rheological properties of sodium alginate in an aqueous system during gelation in relation to supermolecular structures and Ca^{2+} binding," Food Hydrocolloids **23**, 1746–1755 (2009).
[15]P. Lee and M. Rogers, "Effect of calcium source and exposure-time on basic caviar spherification using sodium alginate," Int. J. Gastronomy Food Sci. **1**, 96–100 (2012).
[16]C. Araki and S. Hirase, "Studies on the chemical constitution of agar-agar. Exhaustive mercaptolyses of agar-agar," Bull. Chem. Soc. Jpn. **26**, 463–467 (1953).

[17]V. J. Chapman and D. J. Chapman, "Agar-agar," in *Seaweeds and Their Uses* (Springer, The Netherlands, 1980), pp. 148–193.
[18]Y. Wu, X. Hao, R. Xiao, J. Lin, Z. L. Wu, J. Yin, and J. Qian, "Controllable bending of bi-hydrogel strips with differential swelling," Acta Mech. Solida Sin. **32**, 652–662 (2019).
[19]Z. J. Wang, W. Hong, Z. L. Wu, and Q. Zheng, "Site-specific pre-swelling-directed morphing structures of patterned hydrogels," Angew. Chem., Int. Ed. **56**, 15974–15978 (2017).
[20]Q. Bian, L. Fu, and H. Li, "Engineering shape memory and morphing protein hydrogels based on protein unfolding and folding," Nat. Commun. **13**, 137 (2022).
[21]Z. Xu and J. Fu, "Programmable and reversible 3D-/4D-shape-morphing hydrogels with precisely defined ion coordination," ACS Appl. Mater. Interfaces **12**, 26476–26484 (2020).
[22]H. Hu, C. Huang, M. Galluzzi, Q. Ye, R. Xiao, X. Yu, and X. Du, "Editing the shape morphing of monocomponent natural polysaccharide hydrogel films," Research **2021**, 9786128.
[23]Y. Luo, X. Lin, B. Chen, and X. Wei, "Cell-laden four-dimensional bioprinting using near-infrared-triggered shape-morphing alginate/polydopamine bio-inks," Biofabrication **11**, 045019 (2019).
[24]J. Lai, X. Ye, J. Liu, C. Wang, J. Li, X. Wang, M. Ma, and M. Wang, "4D printing of highly printable and shape morphing hydrogels composed of alginate and methylcellulose," Mater. Des. **205**, 109699 (2021).
[25]A. Banerjee and S. Ganguly, "Mechanical behaviour of alginate film with embedded voids under compression-decompression cycles," Sci. Rep. **9**, 13193 (2019).
[26]M. F. Ashby, *Materials Selection in Mechanical Design*, 5th ed. (Butterworth-Heinemann, Oxford, UK, 2016).
[27]C. Wang, C. Cowen, Z. Zhang, and C. Thomas, "High-speed compression of single alginate microspheres," Chem. Eng. Sci. **60**, 6649–6657 (2005).
[28]E.-S. Chan, T.-K. Lim, W.-P. Voo, R. Pogaku, B. T. Tey, and Z. Zhang, "Effect of formulation of alginate beads on their mechanical behavior and stiffness," Particuology **9**, 228–234 (2011).
[29]C. Ouwerx, N. Velings, M. Mestdagh, and M. Axelos, "Physico-chemical properties and rheology of alginate gel beads formed with various divalent cations," Polym. Gels Networks **6**, 393–408 (1998).
[30]C. K. Kuo and P. X. Ma, "Ionically crosslinked alginate hydrogels as scaffolds for tissue engineering: Part 1. Structure, gelation rate and mechanical properties," Biomaterials **22**, 511–521 (2001).
[31]J. Zhang, C. R. Daubert, and E. A. Foegeding, "Fracture analysis of alginate gels," J. Food Sci. **70**, e425–e431 (2005).
[32]J. Zhang, C. R. Daubert, and E. A. Foegeding, "A proposed strain-hardening mechanism for alginate gels," J. Food Eng. **80**, 157–165 (2007).
[33]Y. Tao, Y. Do, H. Yang, Y.-C. Lee, G. Wang, C. Mondoa, J. Cui, W. Wang, and L. Yao, "Morphlour: Personalized flour-based morphing food induced by dehydration or hydration method," in *Proceedings of the 32nd Annual ACM Symposium on User Interface Software and Technology* (ACM, 2019).
[34]M. Falahati, P. Ahmadvand, S. Safaee, Y.-C. Chang, Z. Lyu, R. Chen, L. Li, and Y. Lin, "Smart polymers and nanocomposites for 3D and 4D printing," Mater. Today **40**, 215–245 (2020).
[35]Y. Tao, Y.-C. Lee, H. Liu, X. Zhang, J. Cui, C. Mondoa, M. Babaei, J. Santillan, G. Wang, D. Luo, D. Liu, H. Yang, Y. Do, L. Sun, W. Wang, T. Zhang, and L. Yao, "Morphing pasta and beyond," Sci. Adv. **7**, eabf4098 (2021).
[36]A. Dick, Y. Gao, B. Bhandari, and S. Prakash, "Influence of drying method and 3D design on the 4D morphing of beef products," Appl. Food Res. **1**, 100017 (2021).
[37]A. Heiden, D. Preninger, L. Lehner, M. Baumgartner, M. Drack, E. Woritzka, D. Schiller, R. Gerstmayr, F. Hartmann, and M. Kaltenbrunner, "3D printing of resilient biogels for omnidirectional and exteroceptive soft actuators," Sci. Rob. **7**, eabk2119 (2022).
[38]A. B. Yeatts, C. N. Gordon, and J. P. Fisher, "Formation of an aggregated alginate construct in a tubular perfusion system," Tissue Eng. Part C **17**, 1171–1178 (2011).
[39]Z. Zhang, R. Zhang, L. Zou, and D. J. McClements, "Protein encapsulation in alginate hydrogel beads: Effect of pH on microgel stability, protein retention and protein release," Food Hydrocolloids **58**, 308–315 (2016).
[40]C. A. Schneider, W. S. Rasband, and K. W. Eliceiri, "NIH Image to ImageJ: 25 years of image analysis," Nature Methods **9**(7), 671–675 (2012).