Bioinspiration & Biomimetics

RECEIVED
15 April 2014

REVISED
4 September 2014

ACCEPTED FOR PUBLICATION
21 October 2014

PUBLISHED
13 May 2015

PAPER

Octopus-like suction cups: from natural to artificial solutions

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Keywords: suction cup, octopus sucker, bio-inspiration, wet attachment, pull-off

Abstract

Octopus suckers are able to attach to all nonporous surfaces and generate a very strong attachment force. The well-known attachment features of this animal result from the softness of the sucker tissues and the surface morphology of the portion of the sucker that is in contact with objects or substrates. Unlike artificial suction cups, octopus suckers are characterized by a series of radial grooves that increase the area subjected to pressure reduction during attachment. In this study, we constructed artificial suction cups with different surface geometries and tested their attachment performances using a pull-off setup. First, smooth suction cups were obtained for casting; then, sucker surfaces were engraved with a laser cutter. As expected, for all the tested cases, the engraving treatment enhanced the attachment performance of the elastomeric suction cups compared with that of the smooth versions. Moreover, the results indicated that the surface geometry with the best attachment performance was the geometry most similar to octopus sucker morphology. The results obtained in this work can be utilized to design artificial suction cups with higher wet attachment performance.

1. Introduction

In recent years, the octopus has been an exceptional source of inspiration due to its soft structure, distributed control system, and ability to change the stiffness of its structures [1–3]. Currently, the attachment features of this incredible animal are of great interest to the scientific community [4–14]. Tramacere *et al* [4] have catalogued octopus sucker features that could facilitate the development of innovative, inspired artificial suction cups. A first biomimetic prototype was built starting from a three-dimensional (3D) reconstruction of magnetic resonance images of real octopus suckers [6], and some octopus-inspired actuation solutions have been suggested that use dielectric elastomers [5, 14] and a shape memory alloy [8]; moreover, an energy-saving system has been presented [7] to obtain efficient attachment. In addition, the sucker coordination in manipulation tasks has been studied [12], and the octopus sucker actuation principle has been modelled [13]. This level of interest in octopus suckers for artificial systems is mainly due to the capability of octopus suckers to attach to all nonporous surfaces and generate a very high negative pressure [15]. Although the octopus has been widely studied, the principles underlying the attachment capability of its suckers are still not completely understood. Octopus suckers, which are muscular hydrostats [16], consist of two portions: the infundibulum (labelled I in figures 1(a), (b)), which is the portion that comes into contact with substrates, and the acetabulum (labelled A in figure 1(b)), which is the portion embedded in the arms of the octopus. As shown in figure 1(b), the acetabulum (please refer to A) is an ellipsoidal hollow structure that communicates with the infundibulum via an orifice (labelled o in figures 1(a), (b)). A recent study [17] on the morphology of *Octopus vulgaris* (the most common species of octopus) revealed a structure in octopus suckers that has never been described: a peculiar protuberance located on the roof of the acetabulum (labelled p in figure 1(b)) that is exactly aligned with the aperture of the orifice.

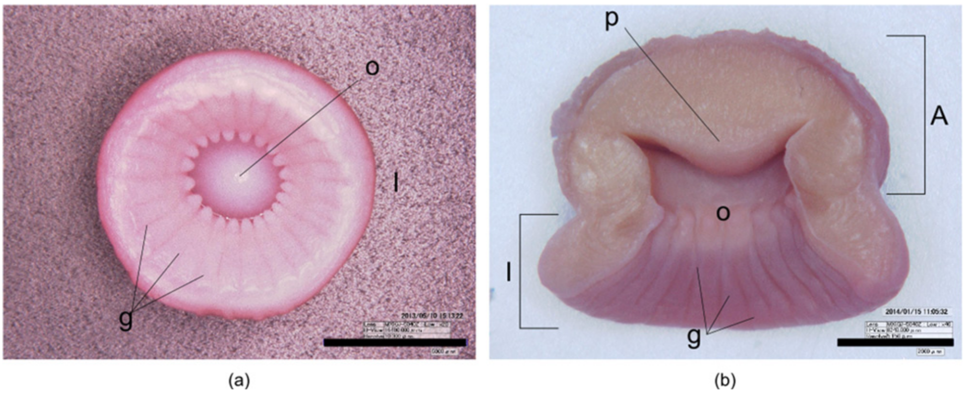

Figure 1. Photographs of *O. vulgaris* sucker. (a) Frontal section of the infundibulum, the externally visible portion of the octopus sucker. The infundibulum is characterized by a series of radial grooves, g, and communicates with the inner part of the sucker via an orifice, o. The scale bar indicates 5 mm. (b) Transverse section. The externally visible portion (infundibulum, I) and the internal portion (acetabulum, A) are labelled. Highlighted: protuberance (p) on the acetabular roof; orifice, o, and grooves, g. The scale bar indicates 2 mm.

In accordance with this finding, an attachment strategy was hypothesized that allows the octopus to remain attached to a substrate or object with minimal energy consumption [17, 18]. Further investigations on the biomechanical features of octopus suckers have been reported [19]. In that work, the authors analysed the surface features and measured the viscoelastic properties of various portions of octopus suckers. The authors noted that the infundibulum consists of very soft tissue, which is as soft as jellyfish jelly ($E \sim 10$ kPa) [19]. The softness of the infundibular portion is critical for the ability of octopus suckers to be absolutely compliant when they contact substrates of various roughnesses. Another aspect that plays an important role in increasing the attachment performance of the sucker in water is the surface morphology of the infundibular portion. As shown previously [19], the infundibular surface of the octopus sucker is characterized by grooves that permit low pressure, which is generated in the acetabular chamber during the active phase of the attachment process; this pressure will be transmitted to almost the entire sucker-substrate interface, thus maximizing the attachment area that is subjected to suction. Because the attachment force is equal to the pressure difference multiplied by the attachment area, the attachment force also increases.

In this study, we aim to investigate the importance and contribution of surface morphology in artificial suction cups, which are similar to octopus suckers, during wet attachment conditions. We have replicated the morphology of the infundibulum of the octopus sucker in artificial devices. We have tested the influence of some artificial infundibulum morphology parameters (e.g., depth and number of grooves, type of material) on attachment performance. We have identified the best morphology for artificial devices, and we have compared this morphology to that of natural suckers. To the best of our knowledge, this study represents the first investigation of the role of surface features in the attachment process of artificial suction cups. Previous studies, which have dealt with wet attachment and bio-inspired solutions, have completely overlooked the contribution of surface morphology to the performance of artificial devices [8, 20–22].

The structure and microstructure of natural suckers are completely different from the smooth surface commonly found in artificial suction cups. This structural difference represents, in our opinion, one of the secrets of the attachment capabilities exhibited by octopus suckers.

2. Materials and methods

2.1. Fabrication of artificial suction cups

Artificial suction cups were produced by casting liquid silicone into a mould. The mould was designed by taking cues from the morphology of actual octopus suckers [6]. The obtained prototypes replicate (in terms of size and proportion) the structure of proximal octopus suckers (the suckers located at the beginning part of the octopus arm with a mean infundibulum diameter of 2 cm). The mould was constructed in resin using a rapid prototyping process (3D System ProJet HD3000, USA) and consists of three components (figure 2(a)).

Three different soft elastomeric materials (Ecoflex 00-30, Ecoflex 00-50, and Dragon Skin 10; Smooth-On, USA) were used to fabricate the artificial suction cups. First, each type of silicone, consisting of a two-component liquid compound, was mixed and then degassed for 5 min in a vacuum oven. Afterward, the silicone was slowly poured into the sealed mould via a syringe (figure 2(b)). The curing phase took 5 h at room temperature. After this phase, the mould was opened, and the suction cups were released. At this point, the infundibular surface of the prototype was completely smooth (figure 2(c), left). To form a

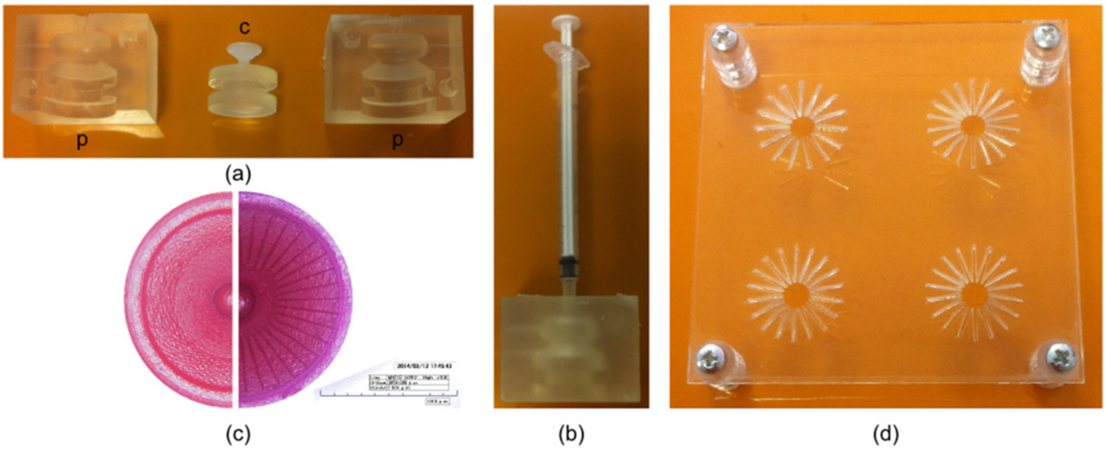

Figure 2. (a) Resin mould used to produce suction cups. The mould consists of three components: two specular external elements, p, and an internal core, c. (b) Sealed mould during liquid silicone pouring. (c) Suction cup prototypes before and after engraving. On the left, the surface of the infundibulum is completely smooth; on the right, the engraved geometry is visible. (d) Plexiglas masks used to flatten the infundibular surface during engraving.

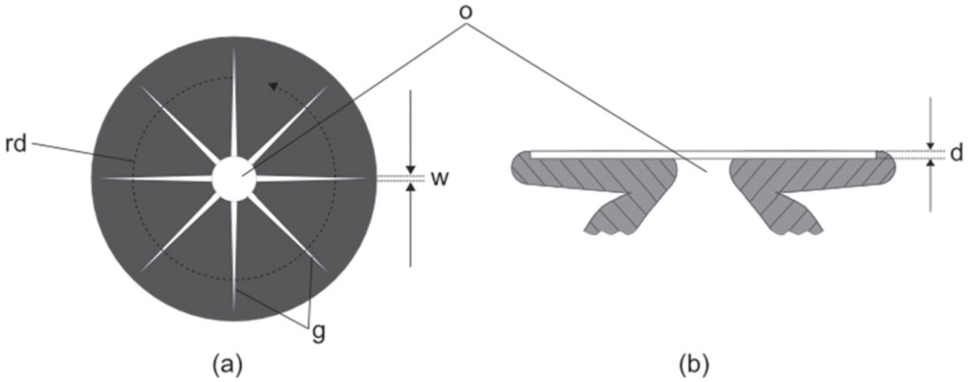

Figure 3. Schematic representation of (a) a frontal view and (b) a transverse section of the infundibulum of a suction cup. Highlighted: grooves, g; orifice, o; radial distribution of grooves, rd; width of groove, w; and depth of groove, d.

network of grooves on the infundibulum, we engraved the silicone materials using a laser cutter (Versalaser VLS 3.50, universal Laser System, USA) (figure 2(c), right). To guarantee that the infundibular portion was completely flat during engraving, a Plexiglas mask with the desired geometry was applied to the suction cup (figure 2(d)).

Suction cups with different surface geometries (N = 18) were produced. Each geometry was characterized by a defined width, radial distribution, and groove depth (w, rd, and d, respectively, in figure 3). Each groove had a triangular shape and radiated outward from the aperture of the orifice to the edge of the rim. Different widths (N = 3) and radial distributions (N = 3) were obtained for the grooves by creating a proper Plexiglas mask for each desired geometry, while different groove depths (N = 2) were obtained by modulating the laser power.

2.2. Pull-off setup and experimental protocol

Pull-off tests were performed to evaluate the attachment performance of artificial suction cups when activated by an external suction system (syringe). The pull-off force in water was measured by applying various pressure differences (between the environment and the internal chamber of the suction cup: 0, 20, 50, and 90 kPa). The setup system consisted of suctioning, anchoring, and pulling-off units (figure 4).

The suctioning unit was responsible for reducing the pressure inside the suction cups. Flexible plastic tubes were used to transmit the suction from the syringe (1.54 cm diameter) (please refer to A in figure 4) to the prototype (please refer to B in figure 4). To measure the difference in the obtained pressure, a differential pressure sensor (26PCCFA2D Honeywell, New Jersey, USA) was placed between the syringe and the prototype using a T-joint (C in figure 4). A syringe pump (D in figure 4) was used to maintain constant suction during the pull-off test without having to continuously and manually pull the syringe. The syringe pump also guaranteed the same pulling phase dynamics for each test. The anchoring unit (figure 4(b)) consisted of a water container (E in figure 4) outfitted with a clamp unit (F in figure 4). The clamp unit fixed the

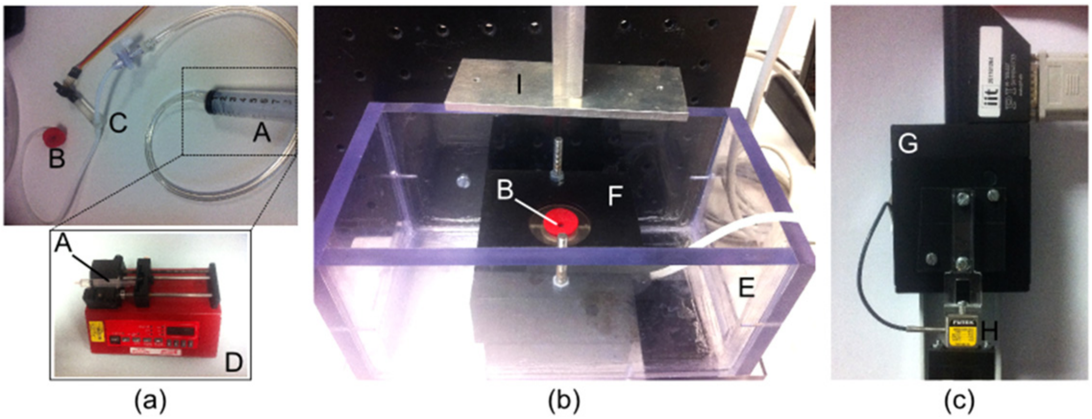

Figure 4. The pull-off setup. (a), (b), and (c) are the suctioning, anchoring, and pulling-off units, respectively. Highlighted: syringe, A; suction cup, B; T-joint, C; syringe pump, D; water container, E; clamp unit, F; translation stage, G; load cell, H; and T-shaped link, I.

suction cup (B in figure 4(b)) to the water container, leaving only the infundibular portion exposed. This configuration prevented any distortion of the suction cup during testing. The pulling-off unit consisted of a micrometre translation stage with a crossed roller bearing (G in figure 4) (M-126.CG1 PI, Karlsruhe, Germany), a uniaxial load cell (H in figure 4) (LSB200 Futek, California, USA), and a T-shaped link (I in figure 4) to anchor the substrate sample. The data from the differential pressure sensor and the load cell were acquired using a data acquisition board (USB6009, National Instruments (NI)) and were recorded using LabVIEW (NI). Data were acquired at a sampling rate of 100 Hz and averaged in windows of 10 samples (final sampling rate of 10 Hz).

Experiments were performed as follows: (i) the substrate was brought into contact with the suction cup (all elements were immersed in water); (ii) the data acquisition for the force and differential pressure was started; (iii) the syringe was pulled to induce suction inside the suction cup, and a constant pressure was maintained during the test; and (iv) the translation stage was activated to induce detachment of the substrate from the suction cup at a velocity of 1 mm s^{-1}; data acquisition was stopped when detachment occurred. The experimental procedure was repeated for each suction cup at each different suction pressure (three times for each suction pressure).

3. Results

3.1. Determination of optimal surface geometry

To quantify the contribution of the surface features to the attachment performance of the suction cups, the maximum force needed to detach the cups from a Plexiglas substrate was measured using a pull-off setup. Artificial suction cups were produced using commercially available soft elastomeric materials (Ecoflex 00-30, Smooth-On, USA) that are widely used in soft robotics [1, 23, 24]. The developed suction cups can be characterized by their surface geometries, which vary in terms of the width, radial distribution, and depth of the grooves (for a total of 18 different combinations). (Please refer to the Materials and Methods section.) To properly evaluate the improvement in attachment performance achieved with the engraved surfaces, the detachment force was measured before (suction cups with a smooth surface) and after engraving (suction cups with engraved surfaces).

Each prototype was tested by applying different pressures (0, 20, 50, and 90 kPa). Both the suction cups with smooth surfaces and the suction cups with engraved surfaces showed comparable mean detachment forces for pressure differences of 20, 50, and 90 kPa. In fact, one-way analysis of variance (ANOVA) of the data revealed p values of 0.95 ($d.f._{total} = 41$; $d.f._{among} = 2$; $d.f._{within} = 39$; $F = 0.06$) and 0.79 ($d.f._{total} = 53$; $d.f._{among} = 2$; $d.f._{within} = 51$; $F = 0.24$) for the smooth and engraved suction cups, respectively. (For the ANOVA of the engraved suction cups, the mean of the attachment performances of all 18 different surface geometries was considered). Both types of suction cups (smooth and engraved) showed a mean increase in detachment force from 0 kPa to 20 kPa, equal to +100% and +145%, for the smooth and engraved suction cups, respectively.

The maximum detachment force of the smooth suction cups was measured to be 2.6 ± 0.2 N. The percent improvements in detachment force for each engraved suction cup are reported in table 1. Improved performance was observed when higher pressure differences were applied (at least 50 kPa) for both the smooth and engraved suction cups. The best performance (with a percentage improvement of 65%) was exhibited by the engraved suction cup with the maximum number of grooves (radial distribution of $1/18\ \pi$), the maximum width of grooves (400 μm), and the smallest depth of grooves (100 μm). Three-way ANOVA was performed considering the different widths, radial distributions, and depths of grooves.

Table 1. Improvements expressed as a percentage (F, mean ± s.d., %) of detachment force and maximum deformation (def, mean ± s.d., mm) of the engraved suction cups. Each measurement was performed on suction cups that varied in the width, radial distribution, and depth of the grooves. Suction cups with different surface morphologies (N = 18) were obtained by combining the three parameters. In detail, three different widths (table rows) and radial distributions (table columns) and two different depths (sub-columns under each column) of grooves were considered (please refer to figure 3).

		Distribution of grooves (rad)					
		1/4 π		2/15 π		1/18 π	
		Depth of groove (μm)		Depth of groove (μm)		Depth of groove (μm)	
Width of groove (μm)		100	200	100	200	100	200
200	F (%)	20 ± 3.0	25 ± 6.0	11 ± 13	20 ± 8.0	48 ± 1.0	42 ± 6.0
	def (mm)	7.5 ± 0.2	7.8 ± 0.1	5.9 ± 0.5	5.9 ± 0.7	8.7 ± 0.0	8.2 ± 0.2
300	F (%)	32 ± 1.0	32 ± 2.0	14 ± 11	20 ± 6.0	55 ± 2.0	53 ± 3.0
	def (mm)	8.2 ± 0.0	7.6 ± 0.5	5.9 ± 0.3	6.3 ± 1.5	8.9 ± 0.3	8.8 ± 0.4
400	F (%)	33 ± 2.0	32 ± 4.0	23 ± 1.0	32 ± 4.0	65 ± 6.0	46 ± 1.0
	def (mm)	8.2 ± 0.5	7.8 ± 0.3	8.1 ± 0.0	7.5 ± 0.2	10.4 ± 0.6	8.8 ± 0.0

Table 2. Morphological details (groove width, w; groove radial distribution, rd; and groove depths, d) (please refer to figure 3) and performance (percentage improvement of detachment force after engraving, F; and maximum deformation, def) of suction cups made from Ecoflex 00-50 and Dragon Skin 10.

	Surface morphological details			Performances	
	w (μm)	rd (rad)	d (μm)	F (%)	def (mm)
Ecoflex 00-50					
geometry 1	200	1/4 π	100	25 π 1	7.4 π 0.1
geometry 2	200	1/18 π	100	32 π 1	8.5 π 0.3
geometry 3	400	1/18 π	100	36 π 1	7.7 π 0.2
geometry 4	400	1/18 π	200	32 π 1	8.6 π 0.3
Dragon Skin 10					
geometry 1	200	1/4 π	100	15 π 1	7.1 π 0.1
geometry 2	200	1/18 π	100	12 π 1	7.3 π 0.2
geometry 3	400	1/18 π	100	16 π 1	7.1 π 0.0
geometry 4	400	1/18 π	200	14 π 1	6.6 π 0.1

The results revealed that the detachment force varied significantly ($p < 0.05$) with variations in these three parameters ($\text{d.f.}_{\text{total}} = 26$; $\text{d.f.}_{\text{within}} = 20$; $\text{d.f.}_{\text{among w}} = 2$, $F_w = 5.2$; $\text{d.f.}_{\text{among rd}} = 2$, $F_{rd} = 6.0$; $\text{d.f.}_{\text{among dg}} = 2$, $F_{dg} = 43.81$; with w, width; rd, radial distribution; and dg, depth of the grooves).

Table 1 summarizes the mean values of the maximum infundibulum deformation before detachment occurred.

As expected, the maximum deformation (10.4 mm) was measured for the suction cup that showed the maximum percent improvement (65%) in detachment force. For the detachment force, a higher degree of deformation occurred when higher pressure differences (at least 50 kPa) were applied. Three-way ANOVA revealed that the maximum deformation varied significantly ($p < 0.05$) with all three tested parameters ($\text{d.f.}_{\text{total}} = 26$; $\text{d.f.}_{\text{within}} = 20$; $\text{d.f.}_{\text{among w}} = 2$, $F_w = 18.5$; $\text{d.f.}_{\text{among rd}} = 2$, $F_{rd} = 13.5$; $\text{d.f.}_{\text{among dg}} = 2$, $F_{dg} = 96.6$; with w, width; rd, radial distribution; and dg, depth of the grooves). In contrast, the smooth suction cups showed a lower maximum deformation of 5.8 ± 0.2 mm.

3.2. Definition of the performance of the optimal material

To understand the role of the suction cup material in the attachment phase, different types of silicone (Ecoflex 00-50 and Dragon Skin 10, Smooth-On, USA) were used. Four surface geometries (please refer to table 2) were selected among the 18 geometries previously analysed. These geometries were chosen to evaluate the influence of each geometrical parameter (width, radial distribution, and depth of grooves) on the performance of the new suction cups during attachment. Pull-off tests using different pressure differences were performed.

3.2.1. Suction cups made of Ecoflex 00-50

The maximum detachment force and infundibulum deformation of the smooth suction cups were 4.3 ± 0.2 N and 5.8 ± 0.2 mm, respectively. The smooth and engraved suction cups showed higher performances when higher pressure differences were applied. The smooth and engraved suction cups exhibited comparable mean detachment forces for pressure differences of 20, 50, and 90 kPa (~4.2 N and ~5.6 N for the smooth and engraved suction cups, respectively). However, the mean percentage increase in the detachment force from 0—20 kPa was +96% and +153% for the smooth and engraved suction cups, respectively. Similar to Ecoflex 00-30, the best engraved suction cup (5.9 N, with a percentage improvement of 36% in detachment force) was the cup with the maximum number of grooves (radial distribution of 1/18 π), the maximum width of grooves (400 μm), and the smallest depth of grooves (100 μm). In contrast to the Ecoflex 00-30 suction cups, increased infundibulum deformation did not occur for the geometry that exhibited a higher detachment force.

All results are reported in table 2.

3.2.2. Suction cups made of dragon skin 10

The maximum detachment force and infundibulum deformation of the smooth suction cups were 8.2 ± 0.4 N and 5.7 ± 0.1 mm, respectively. The smooth and engraved suction cups showed higher performances when higher pressure differences were applied. The smooth suction cups showed a significant difference ($p < 0.05$) in performance for various pressure differences (d.f.$_{\text{total}} = 15$; d.f.$_{\text{among}} = 3$; d.f.$_{\text{within}} = 12$; $F = 149.56$), and the maximum percentage increase in the detachment force from 0 kPa to 90 kPa was +90%. The engraved suction cups exhibited comparable mean detachment forces for pressure differences of 20, 50, and 90 kPa (~9.3 N), and the maximum percentage increase in detachment force from 0—20 kPa was +131%. Similar to previous cases, the best performance in terms of detachment force (9.8 N, with a percentage increase of +16% compared to the smooth version) was observed for the suction cups with the maximum number of grooves (radial distribution of 1/18 π), the maximum width of grooves (400 μm), and the smallest depth of grooves (100 μm). In contrast to the Ecoflex 00-30 cups and similar to the Ecoflex 00-50 cups, the surface geometry that exhibited the highest detachment force for the Dragon Skin 10 cups did not coincide with the surface geometry that showed the highest infundibulum deformation. All results are presented in table 2.

4. Discussion and conclusion

The infundibulum of octopus suckers is characterized by grooves with a mean width of 100 μm, a radial distribution of 1/18—1/10 π, and a depth of 90 μm [19]. These features allow the octopus to maximize the attachment area subjected to suction (generated in the acetabular compartment) and also to maximize the attachment force. The morphology of the infundibulum surface is completely different from the smooth morphology generally found in artificial suction cups. This work presents, for the first time, artificial suction cups inspired by the morphology of octopus suckers. Suction cups were produced using three elastomeric materials with different elastic moduli. Smooth suction cups were produced for casting and were then engraved using a laser cutter. Pull-off tests before (suction cups with smooth surfaces) and after engraving (suction cups with engraved surfaces) were performed to evaluate the attachment performance: the presence of bio-inspired grooves significantly affects the performance, leading to higher adhesive strength, deformability, and thus toughness (energy required for detachment). Although the tests were conducted using various pressure differences, for higher differences, a significant increase in detachment force was not observed. We hypothesize that the absence of such an increase was due to the presence of air bubbles in the suctioning system. Air bubbles indicate the presence of cavitation nuclei, which are very common in real situations. In fact, cavitation is a form of liquid fracture (here water) and would lead to a saturation of the adhesive force similar to the saturation of the strength of a solid. The saturation pressure difference $\Delta p_{\max}$ (the saturation force is $F \approx -\Delta p_{\max} \pi R^2$, where R is the radius of the suction cup; thus, $\Delta p_{\max}$ is not the applied pressure difference) can be estimated according to Laplace's equation. Thus, we find $-\Delta p_{\max} = \frac{2\gamma}{r}$, where γ is the liquid (water) surface tension and r is the characteristic size of the cavitation bubble; according to our observations (best suction cup), we estimate $r \approx 3.5\mu$m. Note that assuming $r \propto R$ would result in a saturation force that scales as $F \propto R$, which is the same scaling as for classical peeling (clearly observed as the applied external pressure difference approaches zero). Finally, it is interesting to note the following analogy: according to fracture mechanics (classical Griffith's case), we could derive a critical negative pressure (fracture strength σ_f) in a form similar to the previous one, namely, $\sigma_f = -\Delta p_{\max} = \frac{2\gamma}{\pi \varepsilon_f r}$, where $\varepsilon_f = \sigma_f / E$ is the fracture strain, E is the Young's modulus of the solid, 2γ is the fracture energy (per unit area), and $2r$ is the crack length. Thus, cracks at the interface between the cup and the substrate would lead to observations similar to those imposed by cavitation.

The softest silicone (Ecoflex 00-30) [19] was used to produce suction cups with 18 different surface geometries. Considering borderline geometries, the attachment performance increased with the number and width of grooves. An unexpected result was obtained for the suction cups with a moderate number of grooves. For these prototypes, we expected better performance than that from the suction cups with fewer grooves. Improved attachment performance with increased groove depth was also observed, except for the geometry with the maximum number of grooves. This behaviour might be explained by the fact that in the suction cups with many grooves, an increase in groove depth leads to an increase in the volume of water present under the grooves. In this situation, the water at the suction cup—substrate interface could affect attachment. The best performance (4.7 N, with a 65% improvement with respect to the smooth version) was obtained using the suction cups with a borderline surface geometry, namely, the maximum number and width of grooves and the smallest groove depth. This surface geometry is very similar to the octopus sucker morphology, except that the width of the grooves in the artificial suction cups is slightly larger than that in natural suckers. Nevertheless, it should be assumed that the surface between two adjacent grooves in an octopus sucker is convex, whereas artificial suction cups have a completely flat surface. Therefore, artificial suction cups may require a larger groove width for better performance. The infundibulum deformation generally followed the detachment force trend (increases with increased width and number of grooves), except for the groove depth. For deeper grooves, the suction cup deformation appeared to be worse.

The suction cups produced with Ecoflex 00-50 showed behaviour equivalent to that of the suction cups produced with Ecoflex 00-30 for the surface geometries tested. Although the best performing engraved suction cup exhibited a higher detachment force (5.9 N), the percentage improvement in detachment force relative to the smooth version (+36%) was considerably lower than that for the suction cups produced with Ecoflex 00-30.

The suction cups produced with Dragon Skin 10 showed comparable detachment forces (~9.5 N) for the four surface geometries tested. However, slightly better performance (9.8 N) was obtained once again by the suction cup with geometry 3 (please refer to table 2). The detachment force of the suction cups produced with Dragon Skin 10 was higher than the force for the suction cups produced with Ecoflex 00-30 and Ecoflex 00-50; however, the percentage improvement relative to the smooth version was significantly lower (~14%).

In light of our results, we can claim that engraving enhances the attachment performance of elastomeric suction cups in all of the tested cases. The best surface morphology for the suction cups using the three different elastomeric materials was the morphology that was most similar to octopus suckers. Greater improvements (compared with the smooth version) in the detachment force were obtained for the suction cups produced with softer materials. In contrast, smaller improvements were obtained for the suction cups produced with harder materials. At the same time, the absolute value of the detachment force was higher for the harder silicones. Future works will be focused on developing suction cups composed of two materials with different degrees of softness for the main structure (harder silicone) and for the artificial infundibulum (thin layer of softer silicone) to mimic the arrangement of tissues and functions in octopus suckers. In fact, different elastic moduli have been observed for different infundibulum portions: deeper tissues (muscles) were found to be harder than superficial tissues (epithelium) [19]. Such suction cups should be able to exploit the intrinsic elasticity of a bi-component system and the surface-engraved morphology of a soft layer at the interface. Additionally, this design could be useful for enhancing the detachment process. In fact, when the pressure inside the suction cup increases, the radial grooves favour the expulsion of water at the interface.

Acknowledgments

This work was supported by the COST Action TD0906 'Biological Adhesives: from Biology to Biomimetics.' N.M.P. is supported by the European Research Council with the following grants: ERC StG Bihsnam, ERC PoC Replica2, and ERC PoC Knotouth, as well as by the European Union and by the Provincia Autonoma di Trento, within the Graphene Flagship.

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