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1	The role of hairs in the adhesion of octopus suckers: a hierarchical peeling
2	approach
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15	
16	Abstract
17	Organisms like the octopus or the clingfish are a precious source of inspiration for the design of
18	innovative adhesive systems based on suction cups, but a complete mechanical description of their
19	attachment process is still lacking. In this paper, we exploit the recent discovery of the presence of
20	hairs in the acetabulum roof of octopus suction cups to revise the current model for its adhesion to
21	the acetabulum wall. We show how this additional feature, which can be considered an example of
22	a hierarchical structure, can lead to an increase of adhesive strength, based on the analysis of the
23	cases of a simple tape and an axisymmetrical membrane. Using peeling theory, we discuss in both
24	cases the influence of hierarchical structure and the resulting variation of contact angles on the

adhesive energy, highlight how an increase in number of hierarchical levels contributes to its
increase, with a corresponding improvement in functionality for the octopus suckers.

27 **1. Introduction**

The Octopus vulgaris is one of the most intelligent animals that lives on Earth. It uses its suckers to perform many functions ([1], [2]). In particular, octopus suckers are able to generate a maximum pressure difference of about 0.27 MPa that can be reached in a few milliseconds [3]. Other animals, such as clingfish, exploit suction cups with a bed of microfibrils or "micropapillae", which are tiny soft protuberances that line the cup perimeter, to better adhere to rough rock surfaces underwater [4]. For this reason, these structures represent a remarkable source of inspiration for designing artificial suction cups or adhesives ([5]–[8]). To develop these artificial devices, the full

35 understanding of the adhesion process and the capability to model it correctly is crucial. In the past, 36 octopus suckers and their interaction with the substrate have been studied mainly by analyzing their arrangement [9] and structure ([10], [11]). In Tramacere et al. [9], a method to identify the suckers 37 in the octopus arm was developed in order to better determine its mechanics through imaging. 38 39 Moreover, in Tramacere et al. [10], three techniques (MRI, ultrasonography, and histology) were used to gain a 3D reconstruction of the sucker (Fig. 1). In this context, the acetabulum protuberance 40 in the acetabulum cavity was discovered for the first time. Experimental studies were also 41 performed to measure the full mechanical properties of the octopus sucker tissues in [11]. 42 Unfortunately, a reliable value of the Poisson ratio remains to be obtained. Work is in progress to 43 resolve this issue. The adhesion of the octopus suckers is achieved by exploiting the pressure 44 45 difference between the external environment, the acetabulum cavity and the infundimbulum cavity (Fig. 1a) [12]. To maintain this pressure difference, the acetabulum roof and the acetabulum wall 46 47 must remain in full contact [10]. More in detail, at the initial stage of adhesion, the infundimbulum is the first part of the sucker in contact with the substrate to form a seal. Then, the acetabular radial 48 muscles contract to reduce the internal pressure in the sucker with respect to the external one. 49 50 Finally, the meridional muscle of the acetabulum contracts to achieve contact between the acetabulum roof and the acetabulum cavity. At this point, all muscles are contracted. When they 51 52 relax, the adhesion is maintained by the adhesive force maintaining the two surfaces in contact (the acetabulum roof and the acetabulum cavity) [13]. Morphological studies show that the latter does 53 54 not present any hairs and can be considered flat.

55 As in other bioadhesion problems, peeling theory has been adopted to describe how these two parts of the octopus suckers delaminate [14]. The first elastic approach developed in the literature in this 56 57 respect was the Kendall model [15], which describes the peeling of a thin elastic tape from a rigid 58 substrate. The main physical quantity that governs the attachment, or the detachment, of the tape 59 is the surface energy γ , which is defined as the energy required to generate a unit area of interface (for a certain crack speed), with Mode I (opening) primary separation mode. In the Kendall model, 60 the force necessary to detach the membrane can be determined by adopting an energy-based 61 criterion, imposing the Griffith's balance between the elastic energy, the adhesive energy and the 62 63 work of the applied load [16]. The peeling force relative to a tape pulled at an angle θ , is thus:

64
$$F = Etw\left(\cos\alpha_0 - 1 + \sqrt{(1 - \cos\alpha_0)^2 + \frac{2\gamma}{Et}}\right)$$
(1)

where *E* is the Young's modulus of the tape, *t* its thickness and *w* its width. Introducing $\hat{F} = F/(Etw)$, where *Etw* represents the force necessary to generate a unit strain in the tape, and $\hat{\gamma} = \gamma/(Et)$, the relation can be written in non-dimensional form:

68
$$\hat{F} = \cos \alpha_0 - 1 + \sqrt{(1 - \cos \alpha_0)^2 + 2\hat{\gamma}}$$
(2)

69 Starting from this approach, a series of more refined models were developed in order to describe 70 various biological mechanisms of adhesion. Among these, the theory of multiple peeling was 71 introduced to model a system of numerous tapes loaded by a single force at a common point [17]. This was used in complex adhesive systems, e.g. to describe the adhesive behaviour of spider web 72 73 anchors [18], [19], [20]. Effects such as tape geometry, viscoelasticity or surface roughness [21], [22] 74 have also been considered, as well as bending stiffness[23]. Moreover, a so-called "hierarchical shear lag model" was introduced to model hierarchical contact splitting occurring in biological 75 76 adhesive structures such as gecko pads [24], [25], which are suitable for active dynamic short-term attachment, and other approaches have considered the effect of pretension in hierarchical 77 structures [26]. These works showed that hierarchical structuring of the surface also leads to the 78 79 reduction of stress concentrations and the appearance of multiple separate peeling fronts, with a 80 resulting increase in adhesive capabilities. These examples indicate the possibility of exploiting 81 various types of structures present in nature for enhanced adhesion in artificial adhesives.

82 The recent discovery of the presence of hairs in the acetabulum roof of the octopus' suckers [27] (Fig. 1) suggests a revision of the model outlined in Tramacere et al. [13]. In particular, the peeling 83 model therein can be improved by adding the additional effect due to the presence of hairs on the 84 flat membrane. This work therefore aims to model the peeling process of a membrane equipped 85 with hierarchical hairs, i.e. to analyse how the hairs affect the peeling force. To do this, we apply 86 87 Yao's approach [28] to the geometry of an axisymmetric membrane, formulating a modified 88 expression for the work of adhesion as a function of the surface energy in a hierarchical structure 89 and deriving the corresponding detachment force of the membrane.

90

91 2. Theoretical model

92 2.1 Hierarchical tape with hairs

We analyse a simple tape with hairs at the interface with the substrate, as shown schematically in
Fig. 2, which we define as "hierarchical", meaning that its adhesive properties depend on structures

95 present at two (or more) different size scales. As a first approximation, hairs are considered to be 96 of the same material of the tape (an incompressible soft material with v = 0.5). Furthermore, they are modelled as flat tapes of thickness t_1 , width w_1 detached length L_1 and contact length l_1 . The 97 distance between two adjacent hairs is ρ along both x and y directions, so that $N = lw/\rho^2$ is the 98 total number of hairs. The hairs form an angle α_1 with the substrate that is considered to be 99 constant, and whose relation to the tape contact angle α_0 is discussed below. During the attachment 100 101 and detachment phases, we do not consider bunching effects of the hairs and possible variation effects in the section of the tape. Equation (1) is valid for a simple tape without hairs. The presence 102 103 of hairs on the tape surface results in an increase of the equivalent surface energy, since there is additional elastic energy stored in the hairs themselves that is "dissipated" as kinetic energy 104 105 released after detachment ([25], [29]). Thus, Eq. (1) remains valid and the surface energy term can 106 be modified to

107
$$\gamma' = \gamma + \gamma_H \tag{3}$$

108 where γ' is the total surface energy, γ the surface energy of the flat tape and γ_H the equivalent 109 surface energy due to the additional elastic energy stored in the hairs. As a first approximation, we 110 neglected the roughness of the substrate. According to previous work [22], this roughness is not 111 expected to influence results significantly, unless it is of the order of the microscopic features (i.e. 112 the hairs) of the adhesive surface, which is not the case considered herein.

Since all hairs are assumed identical, γ_H can be considered homogeneous over the whole contact surface, and can be evaluated as:

115
$$\gamma_H = \frac{l_1 + L_1}{2Ew_1^2 t_1 l_1} P_1^2 \quad (4)$$

where P_1 is the detachment force of a single hair. Using Eq. (1) to compute P_1 , we obtain:

117
$$\gamma_{H} = \frac{Et_{1}}{2} \left(1 + \frac{L_{1}}{l_{1}} \right) \left(\cos \alpha_{1} - 1 + \sqrt{(1 - \cos \alpha_{1})^{2} + \frac{2\gamma}{Et_{1}}} \right)^{2}$$
(5)

118 We can now write Eq. (3) in non-dimensional form:

119
$$\hat{\gamma'} = \hat{\gamma} + \frac{\gamma_H}{Et} = \hat{\gamma} + \frac{t_1}{2t} \left(1 + \frac{L_1}{l_1} \right) \left(\cos \alpha_1 - 1 + \sqrt{(1 - \cos \alpha_1)^2 + \frac{2t}{t_1} \hat{\gamma}} \right)^2 \quad (6)$$

Substituting this expression for the surface energy in Eq. (2), we obtain the modified nondimensional pull-off force as:

. 2

122
$$\hat{F} = \cos \alpha_0 - 1 + \sqrt{(1 - \cos \alpha_0)^2 + 2\hat{\gamma} + \kappa_1 \left(\cos \alpha_1 - 1 + \sqrt{(1 - \cos \alpha_1)^2 + \lambda_1 \hat{\gamma}}\right)^2}$$
(7)

Where $\kappa_1 = \frac{t_1}{2t} \left(1 + \frac{L_1}{L_1}\right)$ and $\lambda_1 = \frac{2t}{t_1}$. Equation (7) thus represents the dimensionless force 123 necessary to detach a rectangular tape equipped with hairs. Notice that the area fraction, i.e. the 124 125 ratio between the contact areas of the tape with/without hairs, respectively, is usually considered close to 1, i.e., the presence of hairs does not entail a reduction/increase of the contact area[25]. 126 To illustrate the resulting behavior, we plot the peeling force \hat{F} in Fig. 3b for various angles ϵ , having 127 chosen the following parameters: $\hat{\gamma}$ = 4·10⁻⁴ , $w = 10^{-2}m$, $l = 10^{-2}m$, $t = 10^{-3}m$, $w_1 = 10^{-5}m$, $l_1 = 10^{-5}m$, $l_2 = 10^{-5}m$, $l_2 = 10^{-5}m$, $l_2 = 10^{-5}m$, $l_3 = 10^{-5}m$, $l_4 = 10^{-5}m$, $l_5 = 10^{-5}m$, 128 $10^{-5}m$, $L_1 = 10^{-5}m$, $t_1 = 10^{-5}m$. As expected, the presence of a hierarchical structure, i.e. of 129 hairs, contributes to an increase of the adhesive properties of the tape for all peeling angles due to 130 the additional stored elastic energy, which is dissipated during delamination, with an increased 131 effect for small angles. The peeling force decreases only slightly for increasing ϵ values. For $\alpha_0 = 0$, 132 and $\alpha_1 = 0$, the tape is sheared parallel to the surface, and the additional dissipated energy due to 133 134 the contribution of the hairs is maximum. Conversely, their decreasing effect when the peeling angle increases and tends to $\pi/2$ is consistent with the qualitative behavior observed in biological 135 adhesion, where the peeling force needs to be maximized mainly for small peeling angles, while 136 facilitated detachment is required at larger angles, to achieve the ON/OFF mechanism necessary, 137 e.g. for motion in animals like geckos or insects like beetles. 138

It should be noted that in Eq.(1) and its derivations, we neglect the effect of the deformation of the substrate. In previous work, the presence of a soft substrate in peeling problems was seen to give rise to an overall increase in the detachment force, due to a wider load distribution at the interface, reducing the load concentration at the peeling line, and a decrease of the local peeling angle [22]. Thus, we expect the soft substrate not to affect the predicted qualitative behavior.

144

145 2.2. Hierarchical axisymmetric membrane

The detachment of a single octopus' sucker can be treated as the peeling of an axisymmetric membrane [13], treated by Afferrante et al. [30], and schematically illustrated in Fig. 4a. The nondimensional force necessary to detach the membrane is

149
$$\hat{F} = \left(\frac{32}{27}\right)^{\frac{1}{4}} (\hat{\gamma})^{\frac{3}{4}} (1+\hat{a}) \qquad (8)$$

150 where \hat{F} and \hat{a} are the dimensionless normal load and detached radius, respectively. Equation (8) 151 predicts a linearly increasing peeling force with the membrane detached radius \hat{a} , i.e. an adhesive 152 membrane can ideally bear an arbitrary load, provided it is large enough. In this case, the modification of γ due to the presence of hairs should be also considered. By inserting Eq. (6) in Eq. 153 (8) we obtain the non-dimensional force necessary to detach the axisymmetric membrane equipped 154 with hairs, although in this case the latter are assumed to be radially distributed, as shown in Fig. 155 4b. Making the same assumptions as in the previous Section, we obtain the detachment force of 156 the axisymmetric membrane as: 157

158
$$\hat{F} = \left(\frac{32}{27}\right)^{\frac{1}{4}} \left(\hat{\gamma} + \kappa_1 \left(\cos\alpha_1 - 1 + \sqrt{(1 - \cos\alpha_1)^2 + \lambda_1 \hat{\gamma}}\right)^2\right)^{3/4} (1 + \hat{a})$$
(9)

The role of the hairs for the axisymmetric membrane can be visualized in Fig. 5. In this case, we plot 159 the peeling force versus the detached radius \hat{a} for $\hat{\gamma} = 4.10^{-4}$, and various values of α_1 . The 160 dependence is linear, but again, the presence of a hierarchical structure implies a considerable 161 162 increase in the adhesive properties of the membrane for a given detached radius. The influence of the hairs on the peeling force decreases as the angle increases, but the \hat{F} vs. \hat{a} curves remain 163 considerably larger than that relative to non-hierarchical case, even for large angles, e.g. $\alpha_1 = 0.4$. 164 This is again consistent with the qualitative behavior observed in biological adhesion, where the 165 166 peeling force needs to be maximized mainly for small peeling angles.

167 It should be noted that in Eq.(1) and its derivations, we neglect the effect of the deformation of the 168 substrate. In previous work [22], the presence of a soft substrate in peeling problems was seen to 169 give rise to an overall increase in the detachment force, due to a wider load distribution at the 170 interface, reducing the load concentration at the peeling line, and a decrease of the local peeling 171 angle. Thus, we expect the soft substrate not to affect the predicted qualitative behaviour.

172

173 2.3 Additional levels of hierarchy

The previous model can be extended to additional levels of hierarchy, as illustrated schematically in
Fig. 3a. In this case, Eq. (3) can be extended as follows:

176
$$\gamma' = \gamma + \gamma_1 + \gamma_2 + \dots + \gamma_n \qquad (10)$$

where γ_1 coincides with the previously introduced γ_H . The total force necessary to detach this type of tape/membrane can be computed as previously, by recursively adding the terms relative to the appropriate hierarchical level. For example, the second level of hierarchy can be described by adding to $\hat{\gamma}_1$ another term of the form

181
$$\hat{\gamma}_2 = \kappa_2 \left(\cos \alpha_2 - 1 + \sqrt{(1 - \cos \alpha_2)^2 + \lambda_2 \hat{\gamma}} \right)^2$$
(11)

where κ_2 , λ_2 and α_2 are analogous to the first level parameters κ_1 , λ_1 and α_1 , respectively. 182 183 Analogous expressions can be written for i > 2. In order to compute the κ_i and λ_i and α_i parameters, 184 it is necessary to consider the geometry (i.e. geometry and contact angles at the various hierarchical 185 levels) of the new system. The approach outlined in the previous sections can then be adopted to determine higher order surface energy values γ_i to the adhesive energy due to the additional 186 hierarchical levels, and the corresponding peeling force. Given the small bending stiffness of the 187 tapes at the various hierarchical levels, the angle variations from one hierarchical level to the next 188 189 are in all cases small. Therefore, the corrections decrease in magnitude for an increasing number of levels, i.e. the adhesive energy and force values do not diverge. This can be seen in results illustrated 190 191 in Fig. 6. Here, we consider as previously a perturbation ϵ on the contact angle from one level to the 192 next, and assume for simplicity that the perturbation is of the same order for each level, i.e. $\cos \alpha_{i+1} = \cos(\alpha_i + \epsilon)$, $\forall i$. Thus, an increase of the hierarchical level also implies an increase in 193 the overall perturbation on the initial peeling angle α_0 . Figures 6a and 6b show the effect of an 194 increasing number of hierarchical levels for the \hat{F} vs. α_0 and \hat{F} vs. \hat{a} plots in the case of a hierarchical 195 tape and a hierarchical axisymmetric membrane, respectively. For 3 levels of hierarchy, at $\alpha_0 = 0.1$ 196 the adhesive force is increased by approximately 6 times with respect to the non-hierarchical case. 197 198 It is apparent that the main increase takes place for the first hierarchical levels, as is clearly visible in Figs. 6c and 6d, where \hat{F} is plotted as a function of the number of hierarchical levels for fixed heta199 and \hat{a} values, again in the case of a hierarchical tape and a hierarchical axisymmetric membrane, 200 201 respectively. We can compute the gain in adhesive force at level *i* by dividing F_i by the force at level 202 *i*-1 (F_{i-1}).

$$Gain = \frac{F_i}{F_{i-1}} \tag{12}$$

Plotting the gain values versus the hierarchical level for the simple tape and the axisymmetric membrane (Fig. 6 e, f), we see that after 2 or 3 levels, there is no further significant gain. Therefore, we can state that 2 or 3 hierarchical levels are sufficient to optimize adhesive force. A further increase in hierarchical levels could be detrimental, since the smallest features would become of the order of the characteristic size of the substrate roughness, leading to a decrease of adhesion

C

[22]. This is consistent with observations on biological adhesive structures found in nature, such as
beetle legs or gecko toes [16][31], which typically display 2 or 3 levels of hierarchy. In the case of
octopus's sucker membranes, hairs appear to be present at most at three levels of hierarchy.

212

213 Conclusions

214 Understanding of the effect of a layer of hairs on the adhesive properties of octopus' suckers is 215 important for the design of artificial suction cups with improved adhesion for various applications, 216 such as smart-skin attachable skin patches [32] or biorobotic adhesive discs [33]. Here, we have 217 evaluated the effect of hierarchical structure, i.e. the presence of hairs, on the adhesion and 218 detachment of a simple tape and of an axisymmetric membrane, in order to gain insight into the 219 adhesion mechanism of octopus' suckers (in particular the detachment of the acetabulum roof from the acetabulum wall). The model is based on a number of simplifying assumptions, e.g. that there is 220 no hair bunching and that the peeling angle does not vary significantly between structures at one 221 222 hierarchical level and those at the next. Furthermore, delamination is assumed to take place from a rigid substrate, whereas the real biological tissue considered is soft and relatively deformable. 223 224 However, these assumptions are not expected to qualitatively modify the analysis herein.

225 Results for the simple tape case indicate that the presence of hairs can improve the adhesive properties by more than 30% at small peeling angles, with the effect decreasing for larger angles. 226 This is consistent with observations on biological adhesion, where typically adhesive forces need to 227 228 be enhanced at small peeling angles. The main parameter determining this increase is the initial 229 detached length of the hairs, which has an upper limit in lengths for which there is an onset of bunching effects. The detachment force for an axisymmetric membrane also increases in the 230 presence of hierarchical structuring. We show that the model can be easily extended to the analysis 231 232 to multiple levels of hierarchy. Here, results indicate that the first hierarchical levels are the ones 233 that contribute more to an increase in adhesive force. In terms of convergence, we find that after the third level of hierarchy there is no longer a significant change in peeling force. 234

This paper provides a possible explanation for the role of the hairs in octopus' suckers, correctly accounting for their role in determining the ON/OFF behavior during adhesion. Currently, further studies are under way to evaluate other possible functions of these hairs (e.g. sensing) that could be fundamental to the octopus functionality. Our work can also help the design of artificial suction cups by providing a model that predicts the potential benefits of a hierarchical surface in terms of
 improved and angle-dependent adhesive properties.

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252 Author Contributions

N.M.P designed the study and supervised the work, G.G. wrote the first draft of the manuscript and
generated diagrams supervised also by F.B.. All the authors finalized the manuscript.

255 Additional Information

256 **Competing Interests:** The authors declare that they have no competing interests.

257 Data availability: The authors declare that the data supporting the findings of this study

are available within the article and its supplementary information files.





Figure 1: a) Schematic of the octopus' sucker: Acetabular Roof (AR), Acetabular Wall (AW) and Infundibulum
(IN). b) Hairs present on the surface of the AR that is attached to the AW during adhesion.





- Figure 2: a) Schematic of the peeling of an elastic tape equipped with hairs; b) schematic of a single
- 266 second -level tape (hair).



Figure 3: a) Schematic of hierarchical levels up to the second order. b) Normalized peeling force vs. peeling angle (Eq. (7)) for different ϵ parameter values and $\hat{\gamma}$ =4·10⁻⁴.



274 Figure 4: a) Schematic of the peeling of an axisymmetric membrane and b) schematic the contact region

between the hairs of the axisymmetric membrane and the substrate.



Figure 5: Peeling force for an axisymmetric membrane vs. detached radius \hat{a} (Eq. (9)) for different α_1 values $(\hat{\gamma} = 4 \cdot 10^{-4})$.



282 Figure 6: a) Normalized peeling force vs. peeling angle for increasing hierarchical levels in the case

of a simple tape ($\varepsilon = 0.05$); b) Normalized peeling force vs normalized detached radius for

increasing hierarchical levels in the case of an axisymmetric membrane ($\varepsilon = 0.05$ and $\alpha_0 = 0.1$). c)

285 Normalized peeling force as a function of number of hierarchical levels in the case of a simple tape

286 ($\varepsilon = 0.05$ and $\alpha_0 = 0.1$). d) Normalized peeling force as a function of number of hierarchical levels in

the case of an axisymmetric membrane (ε = 0.05, \hat{a} =10). e) Plot of the gain (Eq. 12) versus the

288 hierarchical level for the simple tape and f) the axisymmetric membrane.

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