nature portfolio

Peer Review File

Earthworm Inspired Lubricant Self-Pumping Hydrogel with Sustained Lubricity at High Loading

Corresponding Author: Professor Feng Zhou

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
The authors reported the preparation of novel super-lubrication sweaty hydrogels based on multi-level structural design strategy inspired by earthworm. The sweaty hydrogels demonstrate fast surface hydration, reversible pore-closing and pore-opening behavior, along with dynamical lubrication release under mechanical shearing process. After carefully reading and checking the information provided in manuscript, I have to admit that the current sweaty hydrogels has indeed broken the load-bearing limitation of hydrogels-based super-lubricating soft materials, the results are really huge breakthrough in this field (COF<0.01, P> 11 MPa, 100000 cycles). To ensure the accuracy of my judgment, I discussed the results with relevant famous experts in the field, everyone highly recognized the novelty and value of this paper. Overall, the design idea is really novel and results are valuable, I strongly recommend its publication in Nature Communications if the authors address the following comments.

1. The surface of the double-crosslinked hydrogels matrix generated wrinkles after encountering PAA dissociation, this is very interesting. Is this phenomenon caused by swelling-induced mechanics mismatch or other reasons? Please discuss it.
2. The authors used PAA as dissociation agent for achieving super-lubrication (SL) behavior, are other molecules or polymers with carboxyl groups also available for SL? Such as citric acid or carboxymethyl chitosan, please check their possibility.
3. I have carefully checked the results of lubricity evaluation. For the calculation results of interface average contact pressure, when there is no texture, I have no objection to the result. But for textured surfaces in figure 5, the real contact pressure may higher than that of reported values in manuscript, because real contact area is smaller. Authors should discuss this.
4. As stated by the authors, the mechanical strength of the load-bearing phase (double-crosslinked hydrogels matrix) for your layered lubrication hydrogels is high, is there any relative characterizations for supporting its network micro-structure? Such as SEM or AFM?
5. Obviously, the coefficient of friction of your layered lubrication hydrogels is so low (<0.01) at a constant frequency of 1 Hz. I wonder what would happen if further increase the frequency? Will the friction coefficient become lower? Because the interface may enter fluid lubrication regime. Of course, this depends on the sensor accuracy of the friction testing instrument. I suggest authors investigate this at least two extra frequency.
6. As can be seen in Figure 5, the coefficient of friction of your textured layered lubrication hydrogels storing 5 μL lubricant would increase finally. Why? Have the lubricants within the hole been completely consumed? Or the lubricants have lost water and dried up? Please discuss it in details.
7. According to authors' opinion, the appearance of wrinkles or patterns feature on the surface of your layered lubrication hydrogels is benefit to friction-reduction, why? I suggest to add necessary mechanism analysis in supporting information by Figure.
8. There are some grammar mistakes in text, authors should check and revise them carefully.

Reviewer #2

(Remarks to the Author)
Comments for Authors:

Thank you for the opportunity to review your manuscript entitled "Earthworm Inspired Sweaty Hydrogels with Unprecedented

Sustainable Lubricity at High Loading." In this manuscript, the authors synthesize a novel structural soft surface which includes cavities; these cavities are implicated in the robust, very low friction performance that the surfaces achieve under high-pressure loading due to their action as dynamic reservoirs. The surfaces are comprised of hydrogels with softer outer layers (etched by PAA), which are then milled using a laser to created cylindrical divots. Under equilibrium swelling, the cavities close (or partially close), creating the unique surface structures. The combination of the softer outer layer and the lubricant reservoirs allows for robust, very low friction at pressures up to MPa. Overall the work is clearly described and unique; it sets the stage for novel ways of synthesizing hydrogel surface structures using a combination of compositional swelling and manufactured structures. The supplemental videos are helpful and illustrative. All figures are clear and helpful.

While the work is compelling, the authors have used some analogies in the title which detract from the scientific findings and short-change the lubrication mechanism description. In addition more description of the lubrication mechanisms should be included. Thus the following major and minor points should be addressed:

Major Points:

1. Title, etc: The use of the word "sweaty" is not very suitable to describe the mechanism. It is suggested to revise the title, as well as to revise the description of the analogy in the text. The reasons are as follows: A) "sweaty" is an informal word; B) sweating is a 1-way flow of fluid from sweat glands to regulate the temperature of a body, while this lubrication mechanism is the repeated pressurization of water reservoirs through partially-closed pores; C) sweating is active while this lubrication is passive (osmotic potentials change locally under the contact, but ultimately return to the equilibrium swelling state). Rather it is suggested to shift this language toward the idea in Line 255 of "migration" of the fluid due to the pumping action of the migrating contact. Some relevant work on "tribological rehydration" exists in the cartilage community, for example https://doi.org/10.1016/j.joca.2016.09.018
2. Line 89: The use of the analogy of the earthworm seems moderately appropriate, but the way that the synthetic pores resemble the earthworm skin is not clear. It is suggested to add some reference to the specific anatomy, pore spacing, function, etc to strengthen the analogy. In addition, you say on Line 91 that the synthetic glands are "closed." How true is this? Suggest to rephrase this line.
3. One major flaw is the lack of description of the contact area in the friction experiments. The figures are nice, but they obscure the contact so that it cannot be visualized. Is it a flat, a ball? What is the approximate contact area? This is important to report because you are reporting the pressures as a major finding of the work. In addition you implicate the smaller real contact area for lower friction in lines 199-201, but what is the evidence for this? While the structures are not fully collapsing, the contact area is expected to be conformal. Thus, the pores do not contribute significantly to contact area reduction. It is suggested to provide better discussion of this, and to remove the comment about reduced contact area.
4. The lubrication aspects of the paper could be significantly improved by reporting the viscosities of the lubricants, and even calculating a lubrication parameter or showing a lubrication curve. For example in lines 246-248 the viscosities are not given, nor whether all lubricants are expected to be Newtonian. Are either of the polymer solution lubricants shear-thinning? This may help to explain the superior lubrication.
5. Line 342: Jacob Klein is given credit for doing work on the paper, but is not listed as an author. Suggest to correct this oversight.

Minor Points

6. Line 106-107, the composition of the hydrogel is described briefly. Is there a precedent for this particular composition? Why was it chosen?
7. Line 119 you describe the reservoirs as being "filled" but in reality they imbibe the surrounding fluid or supernatant bath. Suggest to rephrase and omit "filled" as it sounds like they are filled independently of the rest of the surface structure, which also swells. This is also confusing in Figure 1B because the blue area and the orange area are both filled with water.
8. Figure S1: This curve clearly shows hyperelastic behavior and hysteresis. What method was used to fit this curve to get the modulus values reported?
9. Figure S22: This figure is very over-simplified. What gives rise to the negative charge? And what is the scale? It is suggested to discuss whether the hydration lubrication or the reservoir pumping is more active in what pressure regimes.
10. Line 187: You report that the friction coefficient increased to 0.006 with very high pressure of 9.9 MPa. Did you assess the sample for damage? Please report the observation.
11. Line 292: "… without any resistance." What does this mean? Resistance to motion is friction, which you report. So there must be some resistance.
12. Grammar and usage are an issue in this manuscript. The more pressing issue is usage. Here are some specific examples:
a. Line 39, etc: "Portholes" is a technical term for windows on a ship; it is suggested to replace all instances of this word. Other more appropriate words would be pores, openings, stoma, or "synthetic glands"
b. Grammar, line 55 "Among," Among what?
c. Readability line 81 add a comma to the large number
d. Grammar line 83 "lubricants-bath condition" is incorrect usage. Suggest to change to " … depends upon the volume of lubricant" or "… depends upon the lubricant bath."
e. Grammar line 90 "so-called" has a hyphen
f. Line 92 & 256: "vividly simulate" is wrong usage and a bit of hype. Suggest to remove the word "vividly."
g. Line 93: Grammar, subject-verb number disagreement
h. Line 100: Grammar, "under confinement environment" is not correct, it needs an article somewhere
i. Grammar lines 100-103, run-on sentence
j. Line 111: Grammar, "wrinkles morphology" should be "wrinkled morphology"

k. Line 216: Typo, “as we soft matter …”
l. Line 248 & 279: Usage, “Amazedly” is incorrect, suggest to change to “Notably” or similar
m. Line 257-261: Run-on sentence
n. Line 280: Grammar, remove “to”

Reviewer #3

(Remarks to the Author)
This is a very interesting paper that describes impressive results. The manuscript, however, is let down by a lack of detail, poor English, lack of references and 'gimicky' presentation.

The manuscript claims to be inspired by the lubrication / sweating of earthworms. However, the manuscript does not contain a single reference to a biological investigation into earthworm epithelia, the secreted aqueous-viscous lubricant and its mechanical or chemical characteristics. The authors claim that this feature has robustness and durability without defining these terms or backing up this claim. Instead they show that the manufacture an artificial earthworm, but the point of making that specific shape remains unclear, the subsequent investigation could have been done with a piece of hydrogel of any shape. This type of gimmicky presentation becomes more evident in Figure 5, where apparently the words robust and lubrication appear, and this is supposedly showing how good the hydrogels performance is.

The manuscript contains a large number of statements where singular and plural are mixed, examples include hydrogels materials, lubricants-composite. Other language issues include the use of the word sustainability, where (probably) sustained is meant. These two have vastly different meanings. A field of study is referred to as a 'cold topic', presumably in contrast to something being a hot topic.

My main point of improvement for the manuscript would be that the materials used and investigations done are poorly described. Based on the information provided, a reader cannot independently build on this work, or verify the results. Recipes are not shared, test regimes are not specified and most importantly: the material is presented as having a coefficient of friction - whilst it is common knowledge that friction refers to a material combination - so there is no point specifying a value if specifics of the counter surface are not provided.

In conclusion: the work is impressive and I would support it being published, but not in its current marketing-style, rather superficial presentation.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
Obviously, the authors provided a detailed response to my comments, especially for the necessary addition of direct measurement of contact area. I noticed some problems that I am concerned, for which that other reviewers have also mentioned them, the authors put in a lot of effort to make revisions. Among, the necessary supplements and modifications to the experimental description section make the technical repeatability of the paper more intuitive, which is great. Moreover, I found the language description level and grammars have significantly improved, and I guess that the authors should have sought help from a professional editing agency. Of course, this is allowed

Overall, I am quite satisfied with the author's revisions. I recommend its publication. By the way, there is one small suggestion. If the authors could add mechanisms description (1-2 sentences) about the robust super-lubricity in the conclusion, it would be even more perfect.

Reviewer #2

(Remarks to the Author)
I have reviewed the first revision of the manuscript now entitled "Earthworm Inspired Lubricant Self-Pumping Hydrogel with Unprecedented Sustained Lubricity at High Loading." The authors have sufficiently addressed all concerns.

Reviewer #3

(Remarks to the Author)
The authors have substantially edited their manuscript in accordance with the reviewers request. Overall I am happy with this manuscript to be published

Reviewer #1 (Remarks to the Author):

General Comment: The authors reported the preparation of novel super-lubrication sweaty hydrogels based on multi-level structural design strategy inspired by earthworm. The sweaty hydrogels demonstrate fast surface hydration, reversible pore-closing and pore-opening behavior, along with dynamical lubrication release under mechanical shearing process. After carefully reading and checking the information provided in manuscript, I have to admit that the current sweaty hydrogels has indeed broken the load-bearing limitation of hydrogels-based super-lubricating soft materials, the results are really huge breakthrough in this field (COF<0.01, P> 11 MPa, 100000 cycles). *To ensure the accuracy of my judgment, I discussed the results with relevant famous experts in the field, everyone highly recognized the novelty and value of this paper. Overall, the design idea is really novel and results are valuable, I strongly recommend its publication in Nature Communications if the authors address the following comments*.

Reply: Thank you for your positive comment and giving high recognition to our work.

Comment 1. The surface of the double-crosslinked hydrogels matrix generated wrinkles after encountering PAA dissociation, this is very interesting. Is this phenomenon caused by swelling-induced mechanics mismatch or other reasons? Please discuss it.

Reply: Thank you for your valuable comment. I completely agree your statement that the patterned wrinkles are generated and caused by the swelling-induced mechanics mismatch. As a response, we have added relative references and simple discussion to support this point.

[55] S. Yang, K. Khare, P. C. Lin, Harnessing surface wrinkle patterns in soft matter, Adv. Func. Mater., 2010, 20(16), 2550-2564.

[56] D. Breid, A. J. Crosby, Effect of stress state on wrinkle morphology, Soft Matter, 2011, 7, 4490-4496.

[57] N. Liu, Q. C. Sun, Z. S. Yang, L. N. Shan, Z. Y. Wang, H. Li, Wrinkled interfaces: taking advantage of anisotropic wrinkling to periodically pattern polymer surfaces, Adv. Sci., 2023, 10, 2207210.

Comment 2. The authors used PAA as dissociation agent for achieving super-lubrication (SL) behavior, are other molecules or polymers with carboxyl groups also available for SL? Such as citric acid or carboxymethyl chitosan, please check their possibility.

Reply: Thank you for your valuable suggestion. According to your suggestion, we have tried other dissociation agents (phytic acid, citric acid and carboxymethyl chitosan) to investigate their possibility for achieving SL behavior at two different normal loads (10 N and 40 N). As shown in Figure R1-R2, even though the friction coefficients both decrease obviously after treating by phytic acid (10 N: 0.0416; 40 N:

0.0492) and citric acid (10 N: 0.0211; 40 N: 0.0258), it is still difficult for them to achieve SL state. By contrast, for carboxymethyl chitosan, the SL state is observed successfully (Figure R3). However, the average friction coefficients in both two normal loads condition (10 N:0.0071; 40 N: 0.0069) are still high than that of PAA (<0.0035).

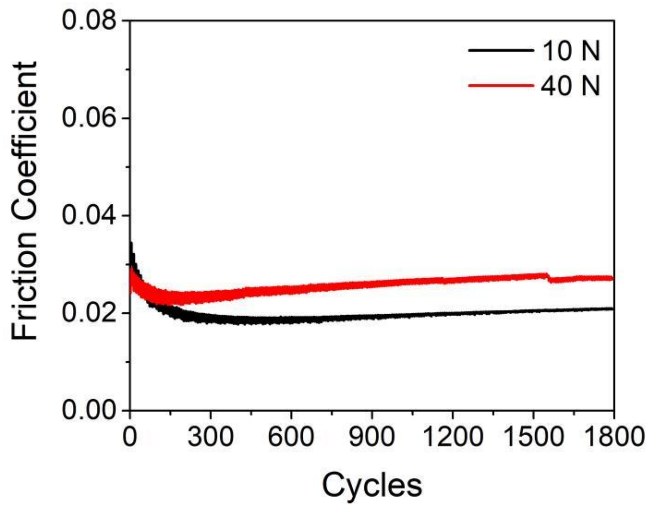

Figure R1. Friction coefficient curves of citric acid-treated high strength DN hydrogel matrix in water (mass concentration: 10%, time: 2 h; frequency: 1 Hz; normal loads: 10 N and 40 N).

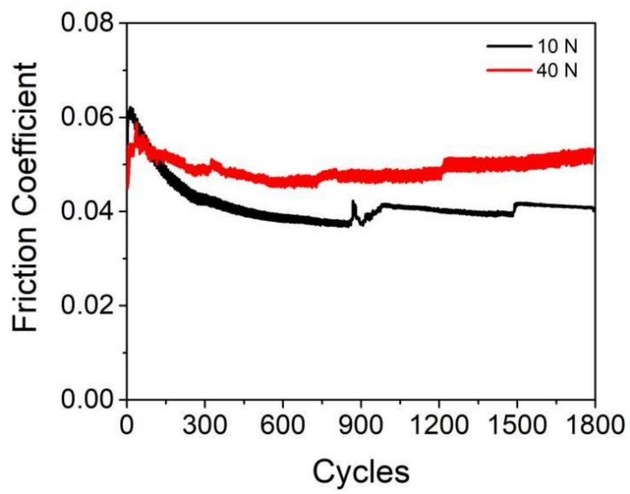

Figure R2. Friction coefficient curves of phytic acid-treated high strength DN hydrogel matrix in water (mass concentration: 10%, time: 2 h; frequency: 1 Hz; normal loads: 10 N and 40 N).

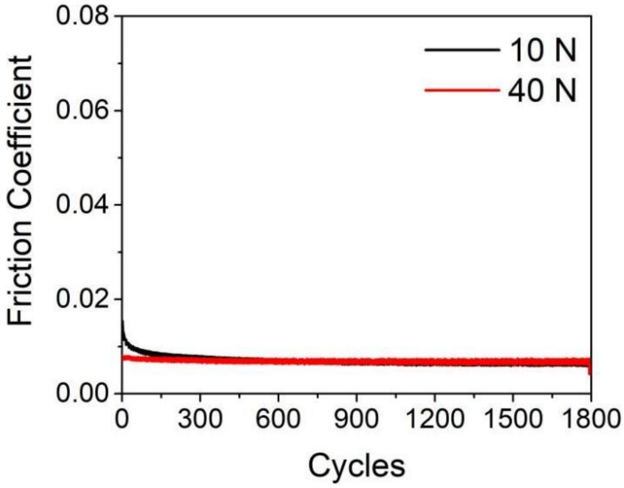

Figure R3. Friction coefficient curves of carboxymethyl chitosan-treated high strength DN hydrogel matrix in water (mass concentration: 5%, time: 2 h; frequency: 1 Hz; normal loads: 10 N and 40 N). Note: due to viscosity and solubility issues, it is difficult to dissolve and obtain a 10% solution.

Comment 3. I have carefully checked the results of lubricity evaluation. For the calculation results of interface average contact pressure, when there is no texture, I have no objection to the result. But for textured surfaces in figure 5, the real contact pressure may higher than that of reported values in manuscript, because real contact area is smaller. Authors should discuss this.

Reply: Thank you for your valuable comment. This comment is similar as that given by the reviewer 2# (third comment). We are sorry for ignoring the textures and wrinkle patterns contributions in calculating the interface average contact pressure. We completely agree your statement that real contact pressure may higher than that of reported values in manuscript, because real contact area is smaller. Due to the opacity of the sample, this poses a great challenge for measuring the contact area. Nevertheless, we still make every effort to solve this problem. As a response, we have built a dynamic observation equipment (Figure R4) for obtaining the real contact areas in varied normal loads (10 N, 20 N, 30 N, 40 N and 45 N, note: the maximum load capacity of the device sensor is 45 N) with considering two key contributions from wrinkle patterns and textured pores. In order to simulate the friction process more accurately, a transparent glass cylinder (d= 3 mm) was used as an indenter to interact with MS-SLH sample. Considering the resolution problem of optical images in the compressed contact area, we used a reflection mode for obtaining contact area of MS-SLH, but a transmission mode for obtaining contact area of SLH. The contact area is calculated by Pixel analysis of optical photographs based on image J software. The Pixel ratio for contact area is defined as Ac (0~1), while the non-contact area is defined as 1-Ac. To ensure statistical error, each optical photograph was divided into four quadrants for analysis (n=4). The apparent contact areas for SLH and MS-SLH samples in manuscript are similar at different normal loads and defined as S_0 (7.065 mm^2), where Ac is 1. The measured contact areas are defined as Sc.

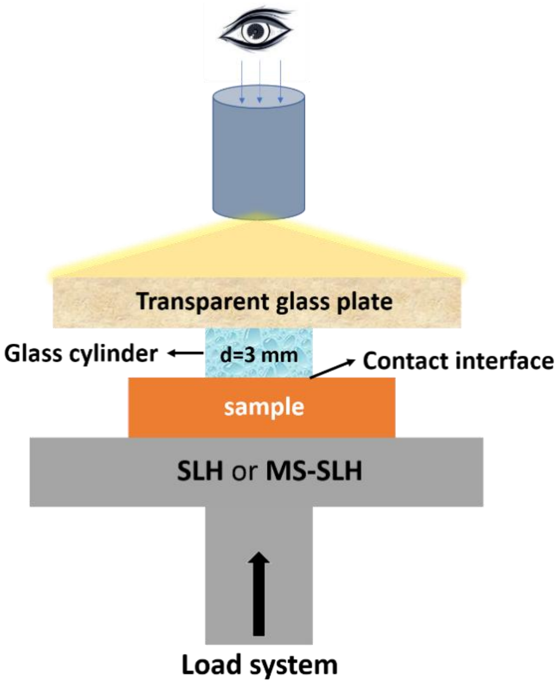

Figure R4. Self-built equipment for observing the real contact state of compressed interface between MS-SLH sample and smooth glass cylinder (d=3 mm) under different normal loads condition.

The obtained optical images of compressed contact interface at different normal loads for SLH sample with wrinkle patterns are shown in Figure R5. Obviously, as the normal load increases, the effective interface contact area ratio (Ac) gradually increases (Figure R6A and R6B). Compared to previous calculation results of interface contact pressures by constant apparent contact area of S_0 (Figure R6C), the calculated contact pressures based on the measured contact area (Sc) at different normal loads increase slightly (Figure R6D).

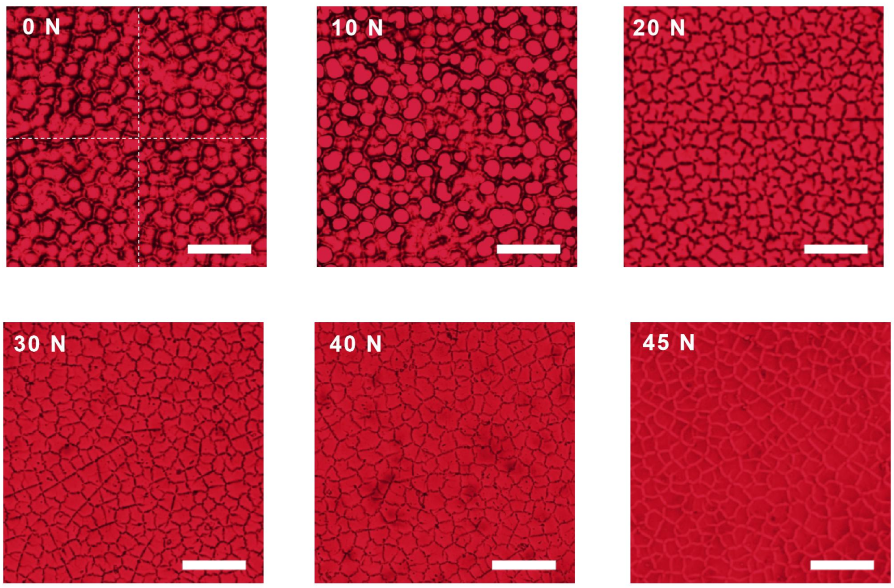

Figure R5. The obtained optical images for compressed contact interface of SLH sample with wrinkle patterns against smooth glass cylinder under different normal loads based on self-built observation equipment (scale bar: 200 μm, the bright red area represents the contact area, the dark black area represents the non-contact area).

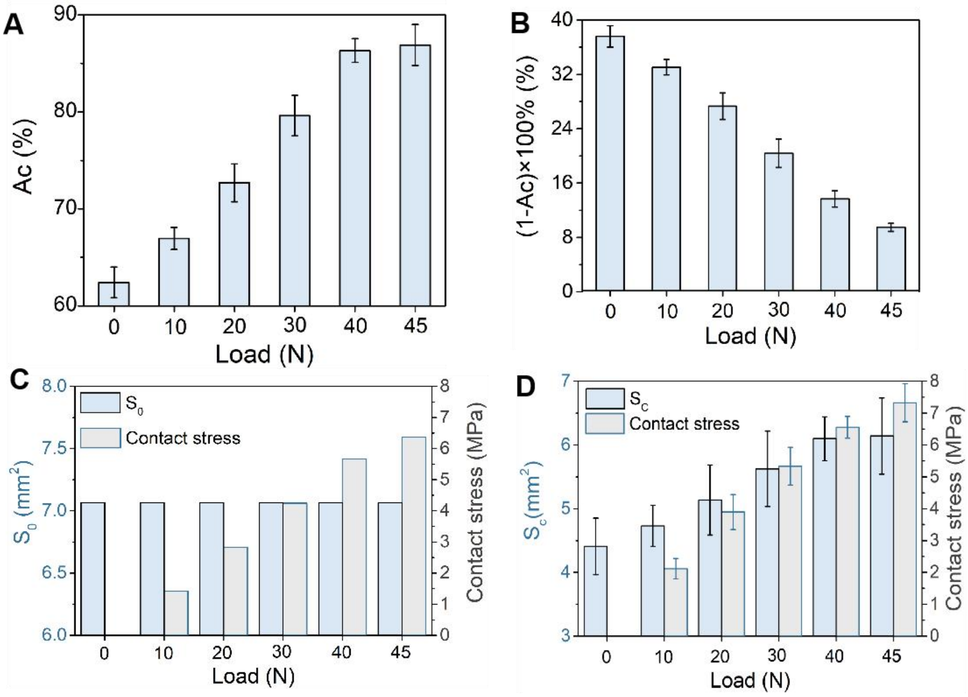

Figure R6. (A) The change of Pixel ratio for contacted area of SLH sample against smooth glass cylinder under different normal loads. (B) The statistical change of Pixel ratio for non-contacted area of SLH sample under different normal loads. (C) The inherent average contact pressures calculated by apparent and constant contact area of S_0 (7.065 mm^2) for SLH sample. (D) The inherent average contact pressures calculated by measured contact area of S_c for SLH sample under different normal loads.

The obtained optical images of compressed contact interface at different normal loads for MS-SLH sample with wrinkle patterns and textured pores are shown in Figure R7. Obviously, as the normal load increases, the wrinkle patterns disappear gradually, while the size of textured pores decreases obviously and keeps unchanged after the normal load increases to 30 N. Correspondingly, the effective interface contact area ratio (Ac) gradually increases and also kept unchanged after the normal load increases to 30 N (Figure R8A and R8B). Compared to previous calculation results of interface contact pressures by constant apparent contact area of S_0 (Figure R8C), the calculated contact pressures based on the measured contact area (Sc) at different normal loads also increase slightly (Figure R8D).

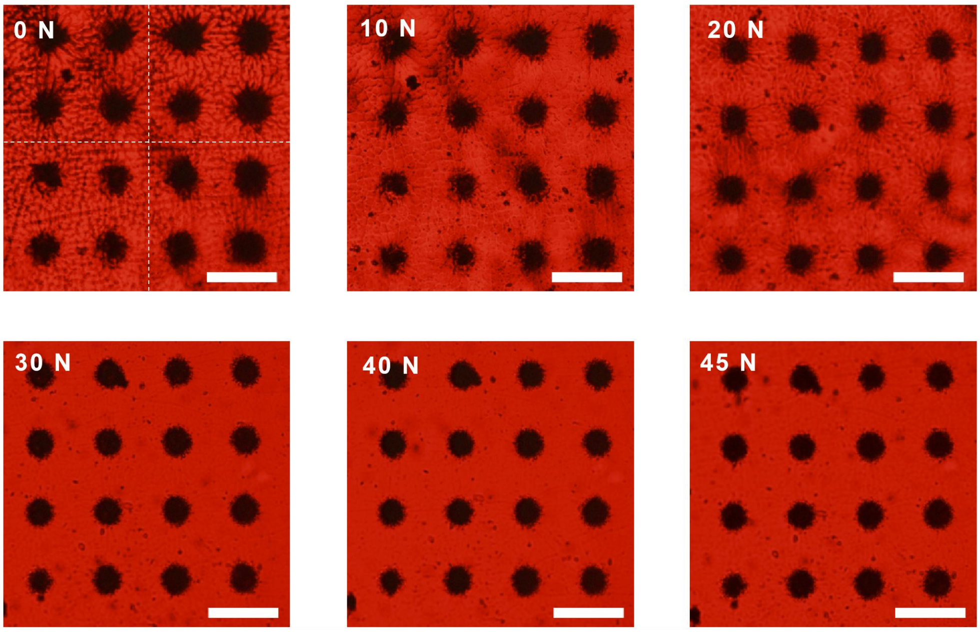

Figure R7. The obtained optical images for compressed contact interface of MS-SLH sample with wrinkle patterns and textured pores against smooth glass cylinder under different normal loads based on self-built observation equipment (scale bar: 200 μm, the bright red area represents the contact area, the dark black area represents the non-contact area).

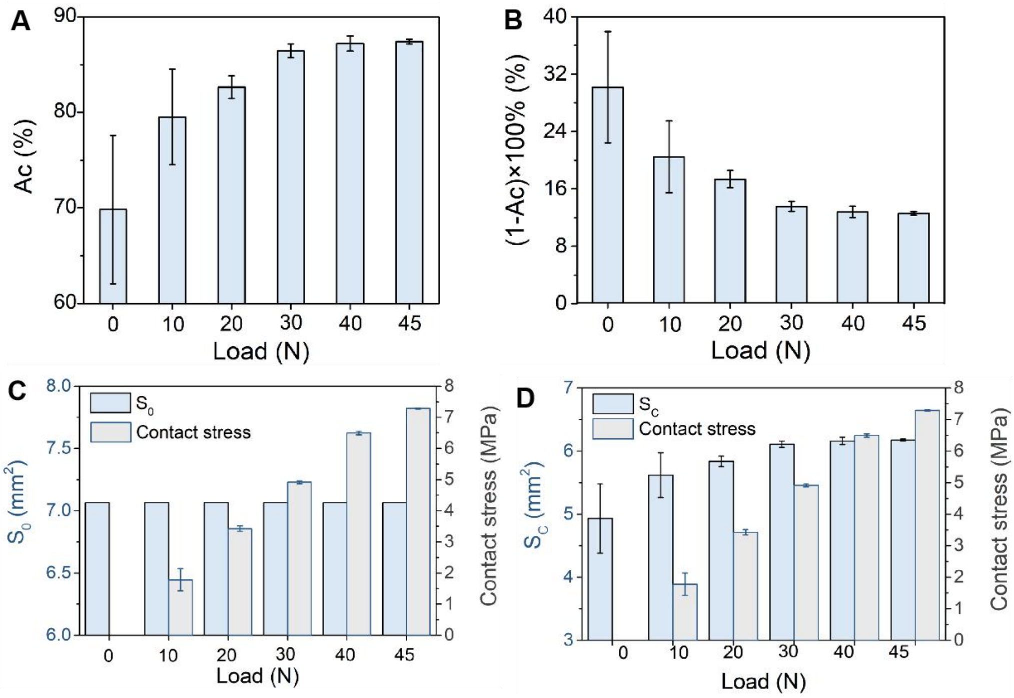

Figure R8. (A) The statistical change of Pixel ratio for contacted area of MS-SLH sample against smooth glass cylinder under different normal loads. (B) The change of Pixel ratio for non-contacted area of MS-SLH sample under different normal loads. (C) The inherent average contact pressures calculated by apparent and constant contact area of S_0 (7.065 mm^2) for MS-SLH sample. (D) The inherent average contact pressures calculated by measured contact area of S_c for MS-SLH sample under different normal loads.

Overall, compared to the calculated contact pressures based on the apparent contact area (S_0), the contributions from wrinkled patterns and textured pores can both effectively reduce the contact area during the friction process, resulting in an obviously increase for the calculated contact pressures. However, for MS-SLH sample, the synergistic evolution of wrinkled patterns and textured pores becomes very complex upon encountering continuous increase of normal loads, making it difficult for us to separate their individual contribution.

Comment 4. As stated by the authors, the mechanical strength of the load-bearing phase (double-crosslinked hydrogels matrix) for your layered lubrication hydrogels is high, is there any relative characterizations for supporting its network micro-structure? Such as SEM or AFM?

Reply: Thank you for your valuable suggestion. As a response, after encountering in situ freezing-drying treatment, we have observed the surface and cross-section morphologies of double-crosslinked hydrogels matrix by FE-SEM and AFM. The results indicate that the network of double-crosslinked hydrogels matrix is very dense (Figure R9-R11), enables to achieve high mechanical strength.

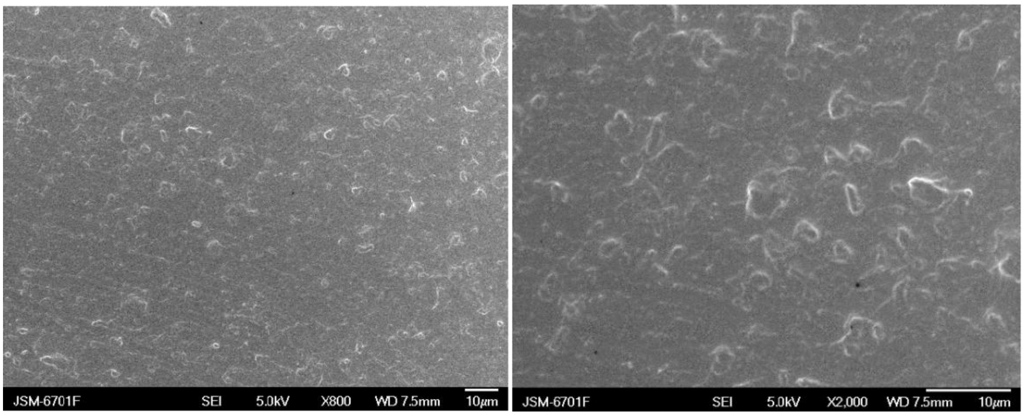

Figure R9. Surface morphology of double-crosslinked hydrogel matrix captured by FE-SEM (the sample was treated by *in situ* freezing-drying treatment).

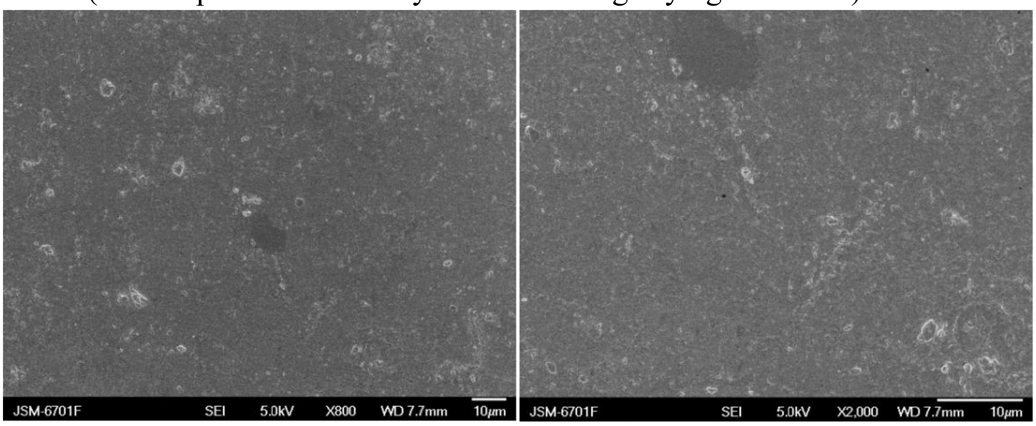

Figure R10. Cross-section morphology of double-crosslinked hydrogel matrix captured by FE-SEM (the sample was treated by *in situ* freezing-drying).

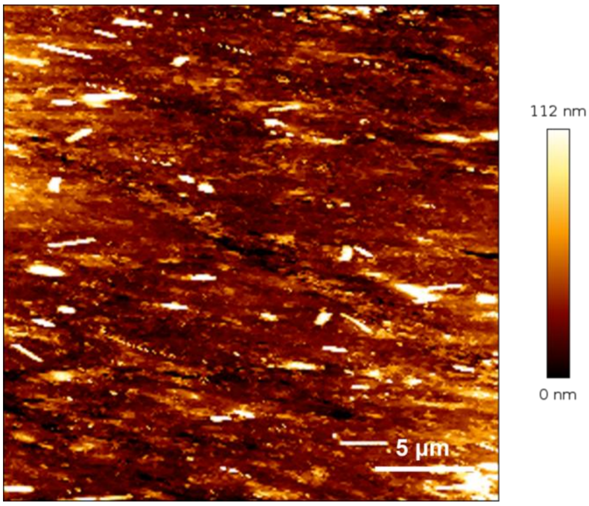

Figure R11. Surface micro-morphology of double-crosslinked hydrogel matrix captured by AFM (the sample was treated by in situ freeze-drying).

Comment 5. Obviously, the coefficient of friction of your layered lubrication hydrogels is so low (<0.01) at a constant frequency of 1 Hz. I wonder what would happen if further increase the frequency? Will the friction coefficient become lower? Because the interface may enter fluid lubrication regime. Of course, this depends on the sensor accuracy of the friction testing instrument. I suggest authors investigate this at least two extra frequency.

Reply: Thank you for your valuable suggestion. In fact, we have investigated the effect of sliding frequency on friction coefficient. The obtained result is shown in Figure R12. As you can see, when we increased the sliding frequency from 1.0 Hz to 1.5 Hz and 2.0 Hz, the friction coefficients of sample decreased significantly. The average friction coefficient of sample at 1.5 Hz sliding frequency in Figure R9 is ~0.0007, while it further reduced to ~0.0003. Indeed, according to the classical lubrication theory, the interface may enter fluid lubrication regime. We were surprised by this result, and discussed it with the machine engineer. After analyzing the data, the engineer indicated that the friction coefficient is too low and achieved the detection limitation of the sensor. The engineer don't recommend us to report the data.

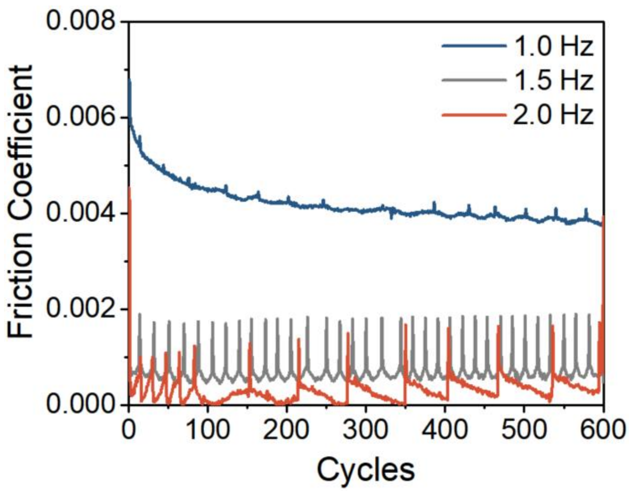

Figure R12. Friction coefficient curves of SLH sample at different sliding frequency

(1.0 Hz, 1.5 Hz and 2.0 Hz).

Comment 6. As can be seen in Figure 5, the coefficient of friction of your textured layered lubrication hydrogels storing 5 μL lubricant would increase finally. Why? Have the lubricants within the hole been completely consumed? Or the lubricants have lost water and dried up? Please discuss it in details.

Reply: Thank you for your comment. The final sharp increase of friction coefficients for MS-SLH samples in Figure 5 is mainly attributed to two factors: the great consumption of lubricants stored within the pores and the complete dehydration of macromolecules lubricants.

Comment 7. According to authors' opinion, the appearance of wrinkles or patterns feature on the surface of your layered lubrication hydrogels is benefit to friction-reduction, why? I suggest to add necessary mechanism analysis in supporting information by Figure.

Reply: Thank you for your comment. Firstly, we would like to clarify that due to the non-regular feature of the wrinkle patterns and the absence of a common divisor between rough peaks, the micro protrusions from the textured patterns will not mechanically bite during the friction process (Figure R13). As shown in Figure R13, we would like to clarify that due to the non-regular feature of the wrinkled pattern and the absence of a common divisor between rough peaks, the micro protrusions from the textured pattern will not mechanically bite during the friction process. The presence of wrinkled patterns will reduce the interface contact area during the friction process. You can find our detail information in reply 3.

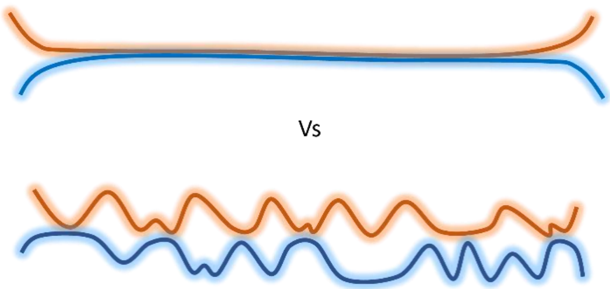

Figure R13. Schematic diagram showing the interface contact states of two pieces of smooth DN hydrogel samples (top) and rough SLH samples in sliding friction process.

Comment 8. There are some grammar mistakes in text, authors should check and revise them carefully.

Reply: Thank you for your suggestion. We have checked the manuscript carefully and polished the English sufficiently by a professional language editing agency.

Reviewer #2 (Remarks to the Author):

Comments for Authors: Thank you for the opportunity to review your manuscript entitled "Earthworm Inspired Sweaty Hydrogels with Unprecedented Sustainable Lubricity at High Loading." In this manuscript, the authors synthesize a novel structural soft surface which includes cavities; these cavities are implicated in the robust, very low friction performance that the surfaces achieve under high-pressure loading due to their action as dynamic reservoirs. The surfaces are comprised of hydrogels with softer outer layers (etched by PAA), which are then milled using a laser to created cylindrical divots. Under equilibrium swelling, the cavities close (or partially close), creating the unique surface structures. The combination of the softer outer layer and the lubricant reservoirs allows for robust, very low friction at pressures up to MPa. *Overall the work is clearly described and unique; it sets the stage for novel ways of synthesizing hydrogel surface structures using a combination of compositional swelling and manufactured structures. The supplemental videos are helpful and illustrative. All figures are clear and helpful.*

While the work is compelling, the authors have used some analogies in the title which detract from the scientific findings and short-change the lubrication mechanism description. In addition more description of the lubrication mechanisms should be included. Thus the following major and minor points should be addressed:

Reply: Thank you for your valuable comments.

Major Points:

Comment 1. Title, etc: The use of the word "sweaty" is not very suitable to describe the mechanism. It is suggested to revise the title, as well as to revise the description of the analogy in the text. The reasons are as follows: A) "sweaty" is an informal word; B) sweating is a 1-way flow of fluid from sweat glands to regulate the temperature of a body, while this lubrication mechanism is the repeated pressurization of water reservoirs through partially-closed pores; C) sweating is active while this lubrication is passive (osmotic potentials change locally under the contact, but ultimately return to the equilibrium swelling state). Rather it is suggested to shift this language toward the idea in Line 255 of "migration" of the fluid due to the pumping action of the migrating contact. Some relevant work on "tribological rehydration" exists in the cartilage community, for example https://doi.org/10.1016/j.joca.2016.09.018
Reply: Thank you for your suggestion. I completely agree with your suggestion, sweaty or sweating is indeed not reasonable for describing the lubrication mechanism of our materials. As a response, we have revised the title and corrected the relevant description. The description of "sweaty" is changed to "Lubricant Self-Pumping". Also, I have read the suggested literature above, and added relative lubrication mechanism description based on"tribological rehydration"effect. This concept vividly

simulates the continuous lubricant migration behavior of epidermis skins at non-pressurized state (Crit. Rev. Environ. Control 1981, 11, 189; J. Agric. Eng. Res. 2001, 79, 239; Soil Tillage Res. 2016, 158, 57), as well as "tribological rehydration" effect of natural cartilage layer for achieving dynamic lubricant supply implemented through mechanical loading and shearing for achieving dynamic lubricant supply (Osteoarthr. Cartilage, 2017, 25(1), 99-107).This novel mechanism could be defined as "lubricant self-pumping". As a response, we have added these key references in main text.

Comment 2. Line 89: The use of the analogy of the earthworm seems moderately appropriate, but the way that the synthetic pores resemble the earthworm skin is not clear. It is suggested to add some reference to the specific anatomy, pore spacing, function, etc to strengthen the analogy. In addition, you say on Line 91 that the synthetic glands are "closed." How true is this? Suggest to rephrase this line.

Reply: Thank you for your good suggestions. This paper mainly imitates the lubrication mechanism of earthworms, rather than the matching structure and secreted substances of earthworms themselves. Earthworms are known to exhibit an extraordinary ability to pass through adhesive soil without inducing stains because of the unique self-lubricating mechanism. This self-lubricating mechanism is based on two factors. One is their sophisticated epidermal glands that can continually secrete water soluble mucus under external mechanical stimuli (such as squeezing and shearing) (Crit. Rev. Environ. Control 1981, 11, 189; J. Agric. Eng. Res. 2001, 79, 239; Soil Tillage Res. 2016, 158, 57). The other is rough skin consisting of macroscopic annuli/microripples which can reduce the friction force and stabilize the secreted mucus to form a thick slippery layer (Journal of Bionic Engineering, 2010, 7(1), 13-18.). For MS-SLH sample of current work, the textured pores mimic the gland of earthworms epidermis, while water or viscous HA/SAA macromolecules lubricants mimic the water soluble mucus form glands of earthworms epidermis. For MS-SLH sample of current work, the wrinkled patterns appeared on surface of soft dissociation lubricating layer mimic the rough micro-structures (annuli and microripple) of earthworms epidermis. Therefore, we believe that the concept of the paper should be reasonable.

To be honest, the glands of earthworms epidermis is almost completely closed. However, the state of textured pore for our MS-SLH sample after swelling equilibrium is partially closed (Figure 4A and Figure 4E). Due to the limitations of the mechanical properties of the soft dissociated lubricating layer, it is currently difficult to manufacture the textured pores with fully enclosed state. However, the lubricity results indicate that partially closed pores do not affect the validation of our concept. We hope the reviewers could kindly understand us. As a response, we have added some relative references to the specific anatomy. Moreover, we have revised previous description from "a closed gland-like lubricant pocket to vividly simulate continuous sweating scenario of skins for achieving dynamic lubricant supply" to "a partially-closed gland-like lubricant pocket to vividly simulate continuous lubricant migration behavior of epidermis skins and cartilage layer for achieving dynamic lubricity maintenance”.

Comment 3. One major flaw is the lack of description of the contact area in the friction experiments. The figures are nice, but they obscure the contact so that it cannot be visualized. Is it a flat, a ball? What is the approximate contact area? This is important to report because you are reporting the pressures as a major finding of the work. In addition you implicate the smaller real contact area for lower friction in lines 199-201, but what is the evidence for this? While the structures are not fully collapsing, the contact area is expected to be conformal. Thus, the pores do not contribute significantly to contact area reduction. It is suggested to provide better discussion of this, and to remove the comment about reduced contact area.

Reply: Thank you for your valuable comments. The friction test conditions are provided in Method (Part 3.7). Specifically, the lubrication feature, load-bearing capacity, and lifespan of the SLH are systematically evaluated by employing typical face-to-face contact mode under with reciprocating sliding style. As a response, we have further revised relative description in main text. The value of average contact pressure is obtained by following the classical formula: P= F/S, where F is the applied normal load, S_0 is the apparent contact area (πr^2=7.065 mm^2).

As shown in Figure R13, we would like to clarify that due to the non-regular feature of the wrinkled pattern and the absence of a common divisor between rough peaks, the micro protrusions from the textured pattern will not mechanically bite during the friction process. The presence of wrinkled patterns will reduce the interface contact area during the friction process.

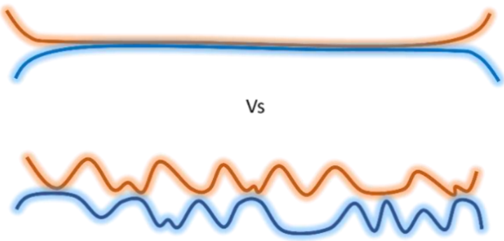

Figure R13. Schematic diagram showing the interface contact states of two pieces of smooth DN hydrogel samples (top) and rough SLH samples in sliding friction process.

For the problem of real contact areas, this comment is similar as that given by the reviewer 1# (third comment). We are sorry for ignoring the texture and patterns contributions in calculating the interface average contact pressure. We completely agree your statement that real contact pressure may higher than that of reported values in manuscript, because real contact area is smaller. Due to the opacity of the sample, this poses a great challenge for measuring the contact area. Nevertheless, we still make every effort to solve this problem. As a response, we have built a dynamic observation equipment (Figure R4) for obtaining the real contact areas in varied normal loads (10 N, 20 N, 30 N, 40 N and 45 N, note: the maximum load capacity of the device sensor is 45 N) with considering two key contributions from wrinkle patterns and textured pores. In order to simulate the friction process more accurately, a transparent glass cylinder (d= 3 mm) was used as an indenter to interact with MS-SLH sample. Considering the resolution problem of optical images in the compressed contact area, we used a reflection mode for obtaining contact area of MS-SLH, but a transmission mode for obtaining contact real SLH. The contact area is calculated by Pixel analysis of optical photographs based on image J software. The Pixel ratio for contact area is defined as Ac (0~1), while the non-contact area is defined as 1-Ac. To ensure statistical error, each optical photograph was divided into four quadrants for analysis (n=4). The apparent contact areas for SLH and MS-SLH samples in manuscript are similar at different normal loads and defined as S_0 (7.065 mm^2), where Ac is 1. The measured contact areas are defined as Sc.

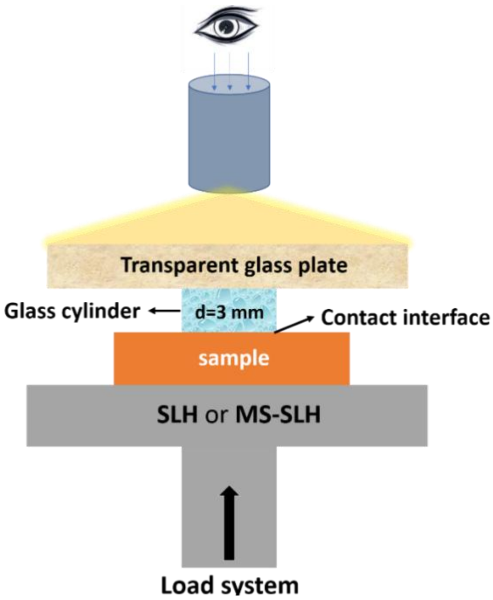

Figure R4. Figure R4. Self-built equipment for observing the real contact state of compressed interface between MS-SLH sample and smooth glass cylinder (d=3 mm) under different normal loads condition.

The obtained optical images of compressed contact interface at different normal loads for SLH samples (wrinkle patterns) are shown in Figure R5. Obviously, as the normal load increases, the effective interface contact area ratio (Ac) gradually increases (Figure R6A and R6B). Compared to previous calculation results of interface contact pressures by constant apparent contact area of S_0 (Figure R6C), the calculated contact pressures based on the measured contact area (Sc) at different normal loads increase slightly (Figure R6D).

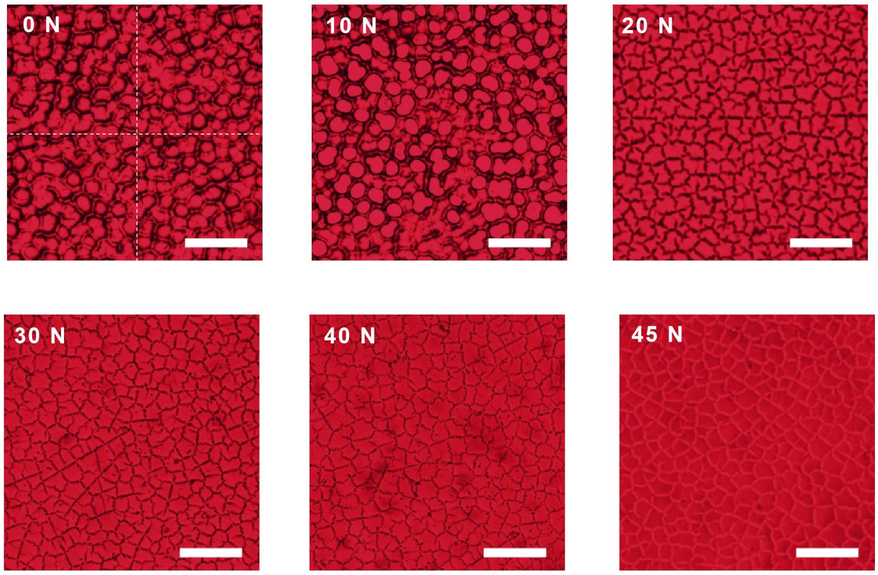

Figure R5. The obtained optical images for compressed contact interface of SLH sample with wrinkle patterns against smooth glass cylinder under different normal loads based on self-built observation equipment (scale bar: 200 μm, the bright red area represents the contact area, the dark black area represents the non-contact area).

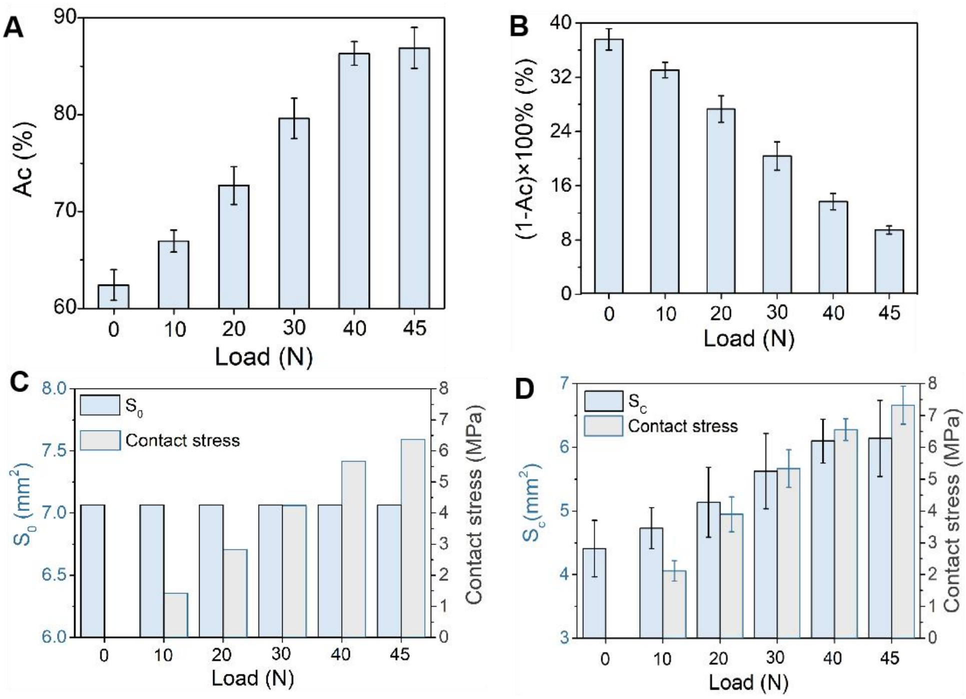

Figure R6. (A) The change of Pixel ratio for contacted area of SLH sample against smooth glass cylinder under different normal loads. (B) The statistical change of Pixel ratio for non-contacted area of SLH sample under different normal loads. (C) The inherent average contact pressures calculated by apparent and constant contact area of S_0 (7.065 mm^2) for SLH sample. (D) The inherent average contact pressures calculated by measured contact area of S_c for SLH sample under different normal loads.

The obtained optical images of compressed contact interface at different normal loads for MS-SLH samples (wrinkle patterns and textured pores) are shown in Figure R7. Obviously, as the normal load increases, the wrinkle patterns disappear gradually, while the size of textured pores decreases obviously and keeps unchanged after the normal load increases to 30 N. Correspondingly, the effective interface contact area ratio (Ac) gradually increases and also kept unchanged after the normal load increases to 30 N (Figure R8A and R8B). Compared to previous calculation results of interface contact pressures by constant apparent contact area of S_0 (Figure R8C), the calculated contact pressures based on the measured contact area (Sc) at different normal loads also increase slightly (Figure R8D).

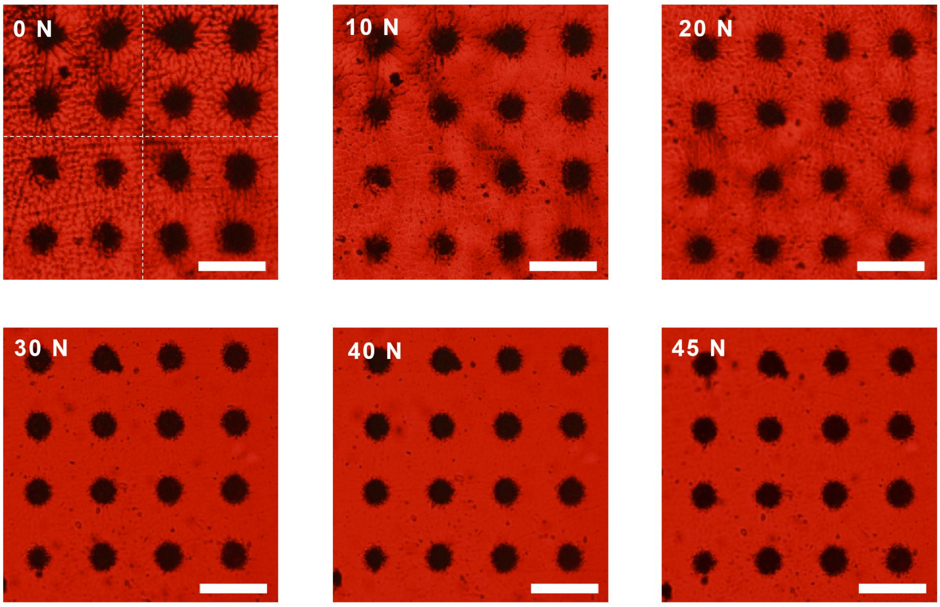

Figure R7. The obtained optical images for compressed contact interface of MS-SLH sample with wrinkle patterns and textured pores against smooth glass cylinder under different normal loads based on self-built observation equipment (scale bar: 200 μm, the bright red area represents the contact area, the dark black area represents the non-contact area).

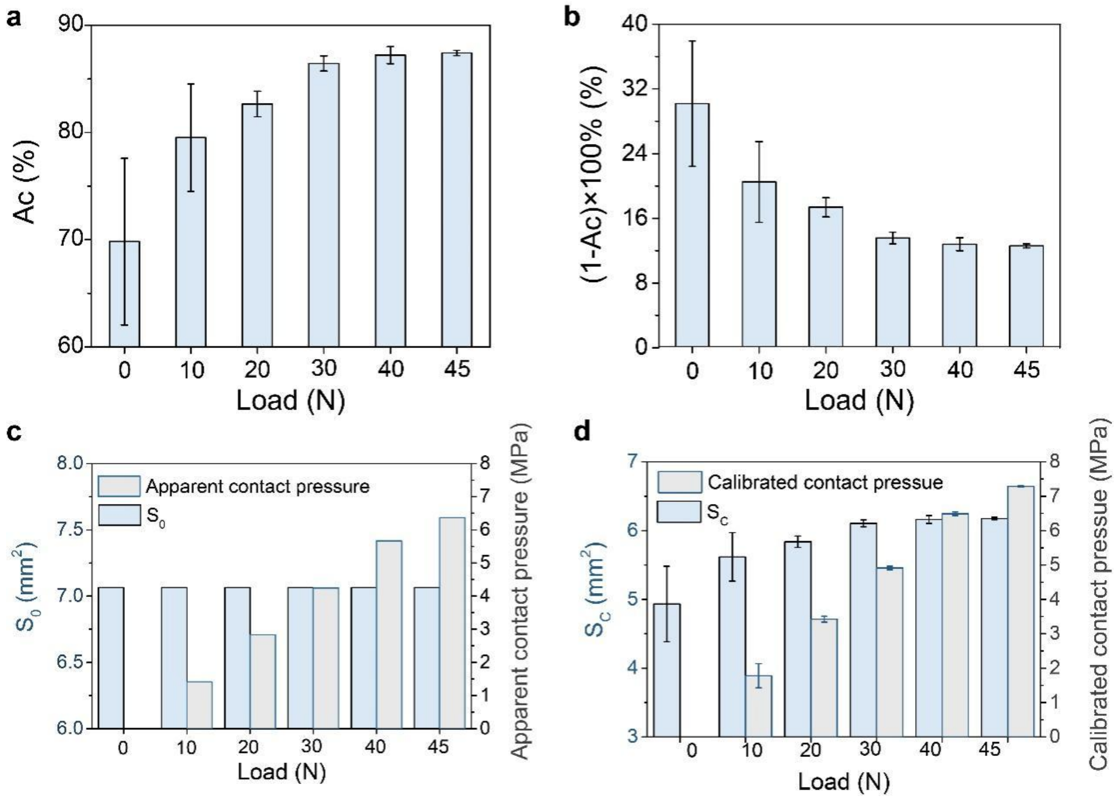

Figure R8. a The statistical change of Pixel ratio for contacted area of MS-SLH sample against smooth glass cylinder under different normal loads. **b** The change of Pixel ratio for non-contacted area of MS-SLH sample under different normal loads. **c** The inherent average contact pressures calculated by apparent and constant contact area of S_0 (7.065 mm^2) for MS-SLH sample. **d** The inherent average contact pressures calculated by measured contact area of S_c for MS-SLH sample under different normal loads.

Overall, compared to the calculated contact pressures based on the apparent contact area (S_0), the contributions from wrinkled patterns and textured pores can both effectively reduce the contact area during the friction process, resulting in an obviously increase for the calculated contact pressures. However, for MS-SLH sample, the synergistic evolution of wrinkled patterns and textured pores becomes very complex upon encountering continuous increase of normal loads, making it difficult for us to separate their individual contribution.

Comment 4. The lubrication aspects of the paper could be significantly improved by reporting the viscosities of the lubricants, and even calculating a lubrication parameter or showing a lubrication curve. For example in lines 246-248 the viscosities are not given, nor whether all lubricants are expected to be Newtonian. Are either of the polymer solution lubricants shear-thinning? This may help to explain the superior lubrication.

Reply: Thank you for your valuable comments. We are sorry for not providing the viscosity parameters of two kinds of lubricants including hyaluronic acid (HA) and sodium alginate (SAA). As a response, the viscosities of HA and SAA are tested by using a rheometer (Figure R14 and Figure R15), as well as the investigation of their shear-thinning feature (Figure R16). In typical case, three shearing rates (0.1 s^{-1}, 100 s^{-1}, 1000 s^{-1}) were employed for viscosity measurement. In order to weaken the influence of shear-thinning effect on measurement, 0.1 s^{-1} may be suitable for obtaining accurate viscosity value. As shown in Figure R14 and Figure R15, the viscosity of hyaluronic acid (HA, 1%) is 4500~7500 mPa.s, while the viscosity of sodium alginate (SAA, 1%) is 1000~2500 mPa.s. These measured results are similar to parameters from agent manufacturer. Compared to low viscosity water, we agree that inherent viscoelasticity of HA and SAA are available for improving the load-bearing capacity. However, as shown in Figure 3G-3I, the load-bearing capacity of sample is already very high, only by employing low viscosity water as lubricant. So, we believe that the viscoelasticity of lubricants themselves is important for load-bearing but not the key factors in current system.

Moreover, the shear-thinning characterizations of the used commercial (A) sodium alginate (SAA, 1%) and (B) hyaluronic acid (HA, 1%) were investigated at wide range of shearing rates (0.05 s^{-1} to 5000 s^{-1}). Obviously, both SAA and HA exhibit significant shear-thinning characteristics, which are crucial for reducing the friction coefficient. This is widely recognized in the field of tribology. However, as shown in Figure 4G and Figure 4J, compared to water, the longer super-lubrication lifetime of MS-SLH sample at limited SAA and HA lubricants amount may be attributed to their extraordinary water retention capacity. By carefully observing the friction coefficient curves in Figure 4J, compared to water, you can find that although the super-lubrication lifetime has become longer for SAA and HA, there is a slight increase in the friction coefficient, which is caused by the viscoelasticity effect of lubricants themselves.

Therefore, we agree that the inherent viscoelasticity and shear-thinning feature are indeed beneficial for improving the load-bearing and friction-reduction functionality, but their contributions may become not obvious under high contact pressure condition. The durability of lubricity with limited lubricants amount (5 μL) is mainly dominated by the continuous self-pumping migration of lubricants along with extraordinary water retention capacity, while the super-low friction feature could be attributed to the synergy from hydration and electrostatic repulsion.

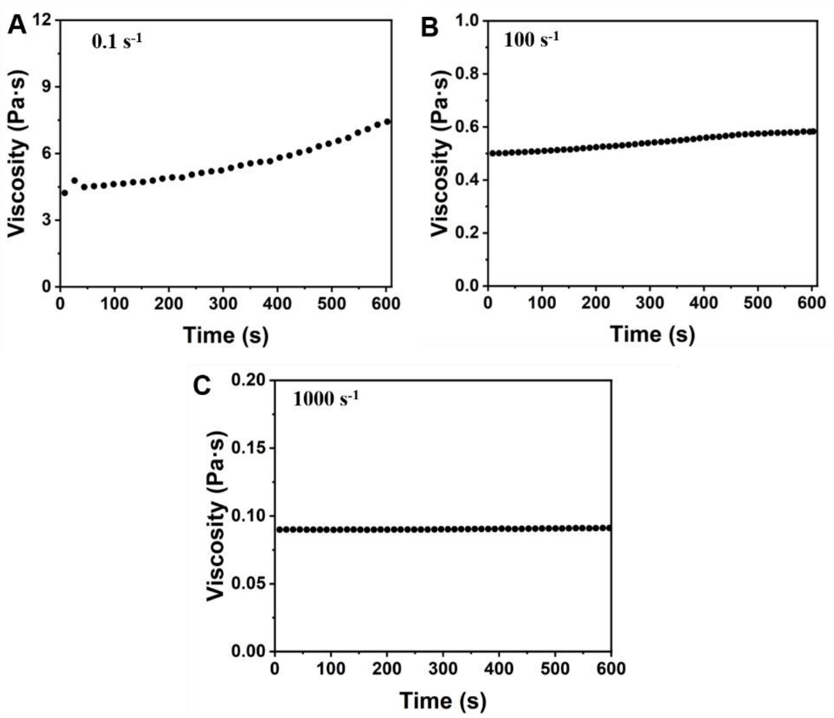

Figure R14. Viscosity measurement of the used commercial hyaluronic acid (HA, 1%) at different shearing rates (0.1 s^{-1}, 100 s^{-1}, 1000 s^{-1}).

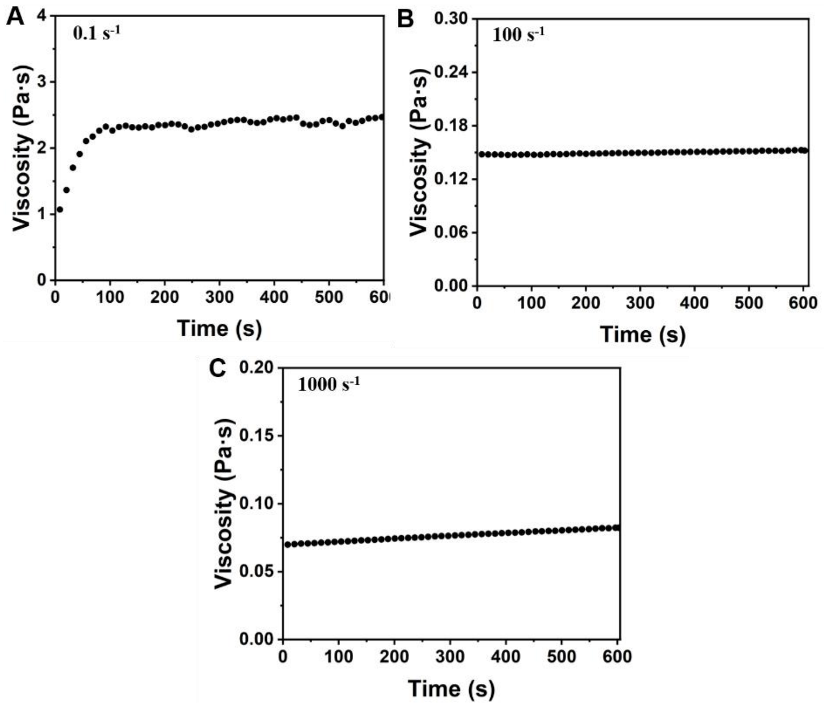

Figure R15. Viscosity measurement of the used commercial sodium alginate (SAA, 1%) at different shearing rates (0.1 s^{-1}, 100 s^{-1}, 1000 s^{-1}).

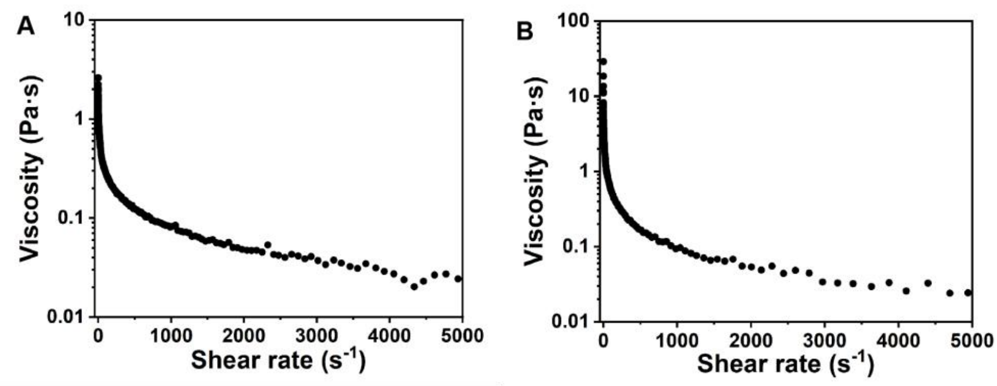

Figure R16. Shear-thinning characterizations of the used commercial lubricants of (A) sodium alginate (SAA, 1%) and (B) hyaluronic acid (HA, 1%) at wide range of shearing rates (0.05 s^{-1} to 5000 s^{-1}).

Comment 5. Line 342: Jacob Klein is given credit for doing work on the paper, but is not listed as an author. Suggest to correct this oversight.
Reply: Thank you for your suggestion. Jacob Klein discussed the experimental results and gave technical guidance. We are very grateful to him and have decided to write his name in authors list. But, when we communicated with the professor Klein before submitting, he insisted on weakening his contribution. So, we can only reflect his contribution in Author Contributions Part. We hope the reviewers can understand.

Minor Points

Comment 6. Line 106-107, the composition of the hydrogel is described briefly. Is there a precedent for this particular composition? Why was it chosen?
Reply: Thank you for your comment. Based on our design concept, we need to find a mechanically tough hydrogel matrix for achieving high load-bearing. On the one hand, the hydrogel matrix must possess high elastic modulus. On the other hand, from the viewpoint of chemistry, the network of hydrogel matrix must be easily and controlled dissociated upon encountering acid-based etchant (-COOH). In order to simultaneously meet these two basic conditions, the double network (DN) hydrogels of poly(acrylamide-acrylic acid)/Fe^{3+} (P(AAm-PAA)/Fe) is chose and prepared as mechanically robust matrix (Figure S1 and Figure S5). Its elastic modulus can achieve as high as 30.45 MPa, while its network contains characteristic group (COOH-Fe-COOH) which could be dissociated by acid.

Comment 7. Line 119 you describe the reservoirs as being "filled" but in reality they imbibe the surrounding fluid or supernatant bath. Suggest to rephrase and omit "filled" as it sounds like they are filled independently of the rest of the surface structure, which also swells. This is also confusing in Figure 1B because the blue area and the orange area are both filled with water.
Reply: Thank you for your suggestion. Indeed, the gland-like pocket imbibes the

surrounding fluid and supernatant bath. However, the swelling degree and water content of the orange area are very low, because the network of DN hydrogel matrix is very dense. As a response, we have changed the original statement from "is in situ filled into" to "spontaneously migrates into".

Comment 8. Figure S1: This curve clearly shows hyperelastic behavior and hysteresis. What method was used to fit this curve to get the modulus values reported?

Reply: Thank you for your comment. Indeed, the curve clearly shows hyperelastic behavior and hysteresis. As a response, we have rechecked the testing data and communicated it with the engineer. The engineer pointed out that the employed normal load (2000 μN) was too high for a soft materials, and the strain range for calculating the elastic modulus value is too far back. As a response, according to the engineer's suggestion, we adopt a reasonable normal load (1000 μN) to measure the elastic modulus again. Then, we obtained the more accurate elastic modulus value (strain range: 20%~80%) of DN high strength hydrogel (Figure R17), its average compression elastic modulus is 30.45 MPa. The newly measured elastic modulus is slightly lower than the previous results.

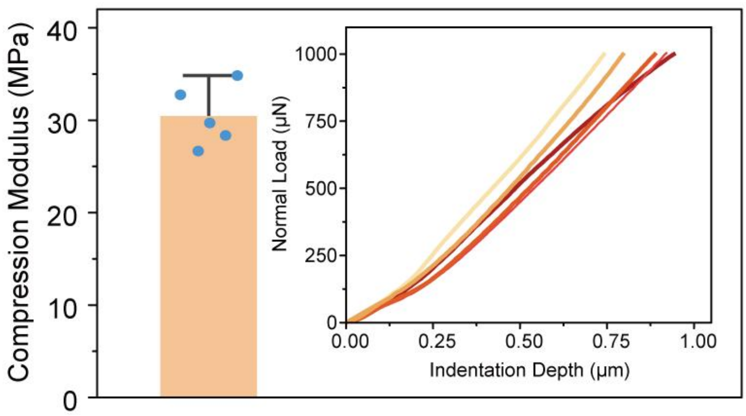

Figure R17. Compression elastic modulus of the mechanically robust DN hydrogel matrix used for generation of dissociation lubrication layer.

Comment 9. Figure S22: This figure is very over-simplified. What gives rise to the negative charge? And what is the scale? It is suggested to discuss whether the hydration lubrication or the reservoir pumping is more active in what pressure regimes.

Reply: Thank you for your comments. The negative charge was generated after the physically cross-linked network (COO^--Fe^{3+}-COO^-) was dissociated by PAA. This could be well proved by the measured results of surface zeta potential (Figure S23). For control sample (DN load-bearing matrix), due to the limited coordination capacity between iron ions (Fe^{3+}) and carboxylate anions (COO^-), a large amount of negative charges remain on the surface of the sample, resulting in a high negative potential value. As we know, PAA is a weak acid, apart from dissociating the physical network (COO^--Fe^{3+}-COO^-) of DN load-bearing matrix, the hydrogen ions (H^+) generated by itself will combine with the COO^- anions within the dissociating network, resulting in a lower surface negative potential value measured compared to the blank DN sample.

Furthermore, as a response, we have added a new Figure R15(Figure S24) to show the robust super-lubrication mechanism of MS-SLH at different contact pressures condition. At low contact pressure condition, the super-lubrication behavior is mainly dominated by hydration lubrication mechanism. By contrast, at high contact pressure condition, the synergy of hydration lubrication mechanism and shearing-induced self-pumping effect well supports the robust super-lubrication behavior.

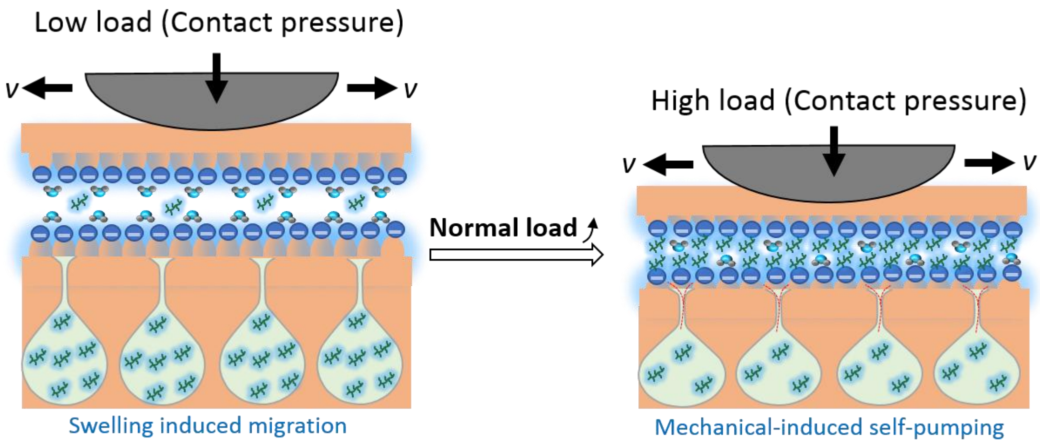

Figure R18. Suggested lubrication mechanism of MS-SLH sample at different normal loads (contact pressure).

Comment 10. Line 187: You report that the friction coefficient increased to 0.006 with very high pressure of 9.9 MPa. Did you assess the sample for damage? Please report the observation.

Reply: Thank you for your comment. In fact, we have assessed the morphology of SLH sample after encountering friction testing, but forgot to show relative results in supporting information. When the applied normal loads increased from 60 N to 70 N, the friction coefficient increased slightly (Figure 3 G). However, there is still no obvious damage on the surface of sample even further increased the normal load to 80 N (Figure R19). Subsequently, we observed the surface morphology of the SLH sample after encountering 100,000 sliding cycles under high normal load of 60 N (8. 47 MPa) (Figure 3I). As shown in Figure R20, compared to original sample (Figure R14A), the surface of SLH sample appeared slight wear, while the non-regular wrinkle patterns feature was still existed, indicating the robustness of the soft lubrication layer.

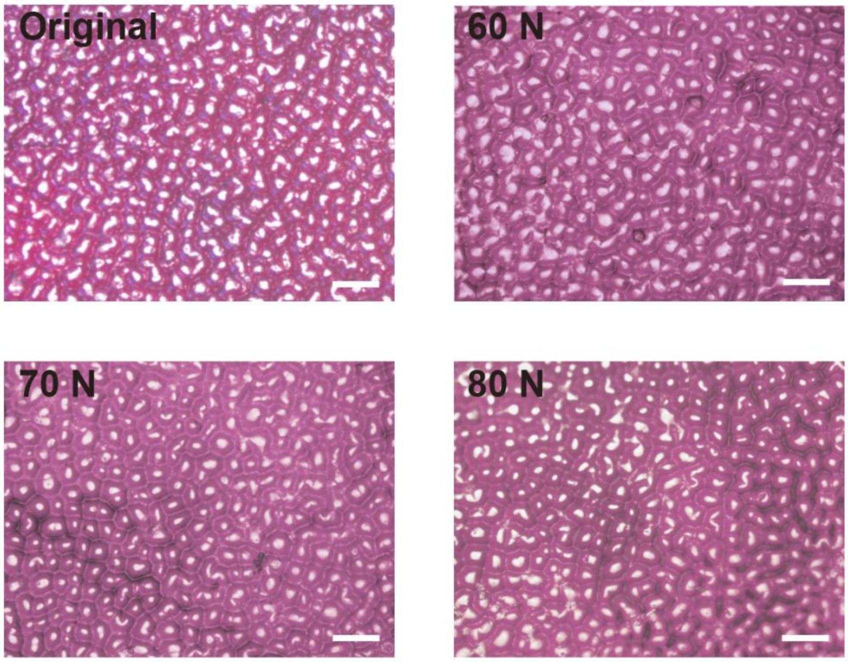

Figure R19. The surface morphology of SLH sample after encountering friction testing under different normal loads (60 N-8.48 MPa; 70 N-9.90 MPa; 80 N-11.32 MPa), the original morphology was used as comparison.

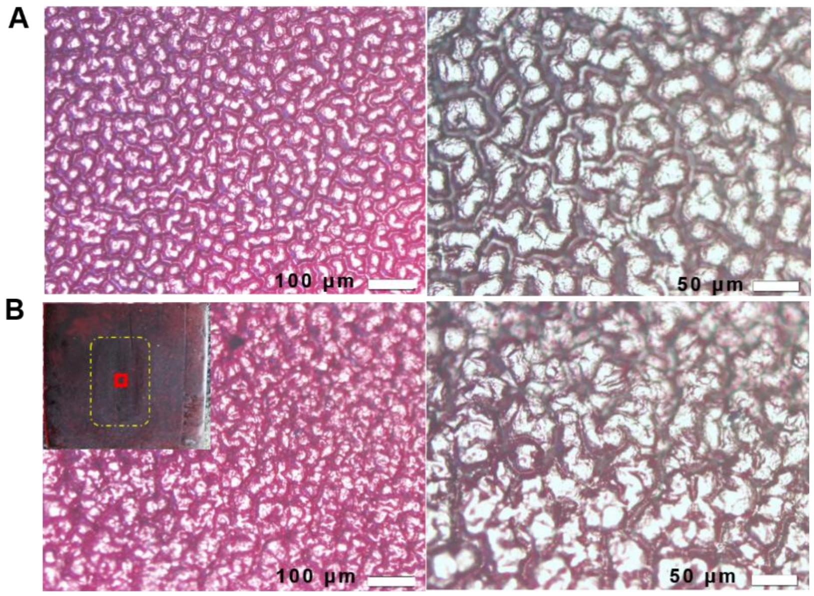

Figure R20. (A) The surface morphology of SLH sample before friction testing and (B) the surface morphology of SLH sample after encountering 100,000 sliding cycles under constant normal load of 60 N (8.48 MPa). The photo inserted in the upper left corner shows the snapshot of the friction testing area (yellow dashed box), the red box represents the center position of the testing area.

Comment 11. Line 292: "… without any resistance." What does this mean? Resistance to motion is friction, which you report. So there must be some resistance.

Reply: Thank you for your comment. We are sorry to make such a low-level mistake.

As a response, we have revised corresponding statement.

Comment 12. Grammar and usage are an issue in this manuscript. The more pressing issue is usage. Here are some specific examples:

a. Line 39, etc: “Portholes” is a technical term for windows on a ship; it is suggested to replace all instances of this word. Other more appropriate words would be pores, openings, stoma, or “synthetic glands”

b. Grammar, line 55 “Among,” Among what?

c. Readability line 81 add a comma to the large number

d. Grammar line 83 “lubricants-bath condition” is incorrect usage. Suggest to change to “ … depends upon the volume of lubricant” or “… depends upon the lubricant bath.”

e. Grammar line 90 “so-called” has a hyphen

f. Line 92 & 256: “vividly simulate” is wrong usage and a bit of hype. Suggest to remove the word “vividly.”

g. Line 93: Grammar, subject-verb number disagreement

h. Line 100: Grammar, “under confinement environment” is not correct, it needs an article somewhere

i. Grammar lines 100-103, run-on sentence

j. Line 111: Grammar, “wrinkles morphology” should be “wrinkled morphology”

k. Line 216: Typo, “as we soft matter …”

l. Line 248 & 279: Usage, “Amazedly” is incorrect, suggest to change to “Notably” or similar

m. Line 257-261: Run-on sentence

n. Line 280: Grammar, remove “to”

Reply: Thank you for your comment. We are sorry to make so many language and grammar mistakes. As a response, we have revised them carefully. Furthermore, we have checked the manuscript carefully and polished the English sufficiently by a professional language editing agency.

Reviewer #3 (Remarks to the Author):

General Comment: This is a very interesting paper that describes impressive results. The manuscript, however, is let down by a lack of detail, poor English, lack of references and 'gimicky' presentation.

In conclusion: the work is impressive and I would support it being published, but not in its current marketing-style, rather superficial presentation

Reply: Thank you for giving us a positive evaluation. Regarding your kind criticism and valuable suggestions, we have tried our best to improve the quality of the manuscript. Especially, we have checked the manuscript carefully and polished the English sufficiently by a professional language editing agency.

Comment: The manuscript claims to be inspired by the lubrication/sweating of earthworms. However, the manuscript does not contain a single reference to a biological investigation into earthworm epithelia, the secreted aqueous-viscous lubricant and its mechanical or chemical characteristics. The authors claim that this feature has robustness and durability without defining these terms or backing up this claim. Instead they show that the manufacture an artificial earthworm, but the point of making that specific shape remains unclear, the subsequent investigation could have been done with a piece of hydrogel of any shape. This type of gimmicky presentation becomes more evident in Figure 5, where apparently the words robust and lubrication appear, and this is supposedly showing how good the hydrogels performance is.

Reply: Thank you for your comments. Your suggestions are really valuable and helpful to us. The core of this paper is to develop mechanically tough hydrogel material with simultaneous high load-bearing, super-lubricating and long-lifetime features, for which its lubrication performance highly surpasses the reported systems (Science, 2021, 374, 212-216; Science, 2020, 370, 6514, 335-338; Figure 3H). The core solution is to find inspiration for material design from nature. This paper mainly imitates the general lubrication mechanism of earthworms, rather than the specific matching structure and secreted substances of earthworms themselves.

Earthworms are known to exhibit an extraordinary ability to pass through adhesive soil without inducing stains because of the unique lubricating mechanism. This unique lubricating mechanism is based on two factors. One is their sophisticated epidermal glands pocket that can continually secrete water soluble mucus (lubricant) under external mechanical stimuli (such as squeezing and shearing), this ensures the lubricity durability (Please see key references to support this: Crit. Rev. Environ. Control 1981, 11, 189; J. Agric. Eng. Res. 2001, 79, 239; Soil Tillage Res. 2016, 158, 57). The other one is rough skin wrinkles consisting of macroscopic annuli/microripples which can reduce the friction force and stabilize the secreted mucus to form a thick slippery layer, this ensures the lubricity robustness (Please see reference to support this: Journal of Bionic Engineering, 2010, 7(1), 13-18; Adv.

Mater. 2018, 30, 1802141). For MS-SLH sample of current work, the textured pores mimic the gland of earthworms epidermis, while water or viscous HA/SAA macromolecules lubricants mimic the water soluble mucus from glands of earthworms epidermis. Furthermore, the wrinkled patterns appeared on surface of soft dissociation lubricating layer of MS-SLH sample well mimic the rough micro-structures (annuli and microripple) of earthworms epidermis. Therefore, we believe that the bionic concept of the paper should be reasonable. Inspired by this bionic concept, we are committed to developing artificial hydrogel lubricating materials with mechanical robustness and lubrication durability. Therefore, our goal is not to create an artificial earthworm, but to learn the natural lubrication mechanism of earthworms and develop high-performance water lubricating materials. The large-scale artificial earthworm prototype (Figure 1C) and series patterns demonstrations (Figure 1D) are only intended to demonstrate the universality and precise controllability of the preparation method. We hope the reviewer can kindly understand us, and we deeply apologize for any misunderstandings caused. Yeah, as you can see, all subsequent performances evaluations of SLH and MS-SLH are based on the sheet-like material. As a response, we have added key references to support the bionic concept from earthworm, as well as the lubricity robustness and durability.

Furthermore, in the demonstrations of Figure 5, the extraordinary performances of high load-bearing capacity, ultra-low friction coefficient, and ultra-long lubricity lifetime for the MS-SLH sample fixed on the slide rail are key focuses, while the two characteristic fonts of "lubrication" and "durability" are just used as LED light indicators for fancy display, and they do not have special significance. The reviewer may have misunderstood this, and we apologize for any problem caused.

Comment: The manuscript contains a large number of statements where singular and plural are mixed, examples include hydrogels materials, lubricants-composite. Other language issues include the use of the word sustainability, where (probably) sustained is meant. These two have vastly different meanings. A field of study is referred to as a 'cold topic', presumably in contrast to something being a hot topic.

Reply: Thank you for your valuable suggestions. We have discussed the reasonable usage problem of singular and complex of hydrogel or composite with experts in this field. As a general category of materials, we should use the plural, while as a specific term in this work, we should use the singular. As a response, we have revised them carefully. Furthermore, we are sorry for making grammar mistakes, we have corrected the wrong word usage from "sustainable" to "sustained. Besides, we have revised the inappropriate expression from "cold topic" to "urgent research topic".

Comment: My main point of improvement for the manuscript would be that the materials used and investigations done are poorly described. Based on the information provided, a reader cannot independently build on this work, or verify the results. Recipes are not shared, test regimes are not specified and most importantly: the material is presented as having a coefficient of friction - whilst it is common knowledge that friction refers to a material combination - so there is no point specifying a value if specifics of the counter surface are not provided.

Reply: Thank you for your valuable comments. In fact, the materials information and experiments parameters have been provided in Supporting Information. We may ignore to introduce some specific details in main text, we are sorry to make inconvenience for your reading. Also, I speculate that the Anna Patterson may forgot to send the supporting information to you, and we apologize for this. Also, the friction testing parameters, counter material and sliding mode were provided in Part 2.6 of supporting information. As a response, we have further checked and revised the supporting information, as well as added necessary information in Main text, to make the reproduction easier for others. We sincerely appreciate the valuable suggestions provided by the reviewer, really thank you very much.

REVIEWERS' COMMENTS

Reviewer #1 (Remarks to the Author):

Obviously, the authors provided a detailed response to my comments, especially for the necessary addition of direct measurement of contact area. I noticed some problems that I am concerned, for which that other reviewers have also mentioned them, the authors put in a lot of effort to make revisions. Among, the necessary supplements and modifications to the experimental description section make the technical repeatability of the paper more intuitive, which is great. Moreover, I found the language description level and grammars have significantly improved, and I guess that the authors should have sought help from a professional editing agency. Of course, this is allowed

Overall, I am quite satisfied with the author's revisions. I recommend its publication. By the way, there is one small suggestion. If the authors could add mechanisms description (1-2 sentences) about the robust super-lubricity in the conclusion, it would be even more perfect.

Response: Thank you for your positive comment and giving high recognition to our work. As a response, we have added lubrication mechanisms description in discussion part.

Reviewer #2 (Remarks to the Author):

I have reviewed the first revision of the manuscript now entitled "Earthworm Inspired Lubricant Self-Pumping Hydrogel with Unprecedented Sustained Lubricity at High Loading." The authors have sufficiently addressed all concerns.

Response: Thank you for your high recognition to our revision. As a response, we have changed the title according to your suggestion.

Reviewer #3 (Remarks to the Author):

The authors have substantially edited their manuscript in accordance with the reviewers request. Overall I am happy with this manuscript to be published.