Reviewer #2

The manuscript by Liu and colleagues concerns the fabrication of novel and flexible SERS substrates for the detection of water pollutants.

The topic is interesting, but in order to make it appealing for this journal (and why they selected this journal) they should emphasize the importance/advantages of using a polymer for the fabrication of such substrates. More in general, the following are the changes I suggest to improve the overall quality of the manuscript.

- Introduction: even though many readers interested in SERS possibly do not need any explanation, the introduction must introduce SERS and the concept of SERS sensors, especially for the average readers. Please try to explain in a more fluent and precise way the concept of SERS, the different mechanisms that generate such enhancement, and also the meaning characteristics of SERS sensors. In addition, please try to cite the articles already published on strictly related examples of SERS uses or substrates fabrication. As an example, it is possible to find works on the use of nanoimprint lithography to produce SERS substrates (since Nanoimprint Lithography of Al Nanovoids for Deep-UV SERS Tao Ding, Daniel O. Sigle, Lars O. Herrmann, Daniel Wolverson, and Jeremy J. Baumberg, ACS Applied Materials & Interfaces 2014 6 (20), 17358-17363), some of them even using sylgard 184 (M Gomez, S Kadkhodazadeh, M Lazzari, Surface enhanced Raman scattering (SERS) in the visible range on scalable aluminum-coated platforms, Chem. Commun., 54 (2018), pp. 10638-10641) to produce flexible SERS substrates (A. Alyami, A.J. Quinn, D. Iacopino, Flexible and transparent surface enhanced Raman scattering (SERS)-active Ag NPs/PDMS composites for in-situ detection of food contaminants, Talanta, 201 (2019), pp. 58-64; Das, A., Pant, U., Cao, C. *et al.* Fabrication of plasmonic nanopyramidal array as flexible SERS substrate for biosensing application. *Nano Res.* (2022). https://doi.org/10.1007/s12274-022-4745-0).

Response: Thank you for the suggestions. There are also some different procedures of SERS substrate, such as nanoimprint lithography and sylgard 184. Ding *et al.* used composite nanoimprint lithography of etched polymer/silica opal films with electron beam evaporation, to fabricate a high-performance sensing substrate for UV-SERS [12]. The UV-SERS performance of DNA base adenine had been enhanced, revealed a well-matching with the UV plasmonic optical signatures and simulations. Moreover, Anindita Das ***et al.*** applied nanoimprint lithography on flexible SERS substrate through electron beam lithography (EBL), which was utilized as a master template and the mold was later replicated via nanoimprinting process to prepare gold-coated polymer nanopyramid three-dimensional (3D) SERS substrate [13]**.** The intense electric field confinement at nanotips and four edges of gold-coated polymer nanopyramid enhanced the Raman signal of probe molecules, which means Rhodamine 6G with a limit of detection down to $3.\mathbf{3} \times 10^{-9}$ M was achieved. Furthermore, Manuel Gómez ***et al****.* presented the potential of combining plasmonic Bragg grating structure, which obtained a rough Al layer to tune SERS enhancement in the visible region [14]**.** The potential structure with excellent analytical reproducibility and amazing enhancement factors (10^{7}-10^{9}) are produced by the scalable procedure, which means it is corresponding to the development of producing sustainable SERS substrates. Abeer Alyami ***et al.***

also fabricated some flexible, sensitive and enable on site detection substrates via Ag NPs/PDMS composites [15]. The Ag NPs/PDMS composites were obtained by self-assembly of Ag nanoparticle solutions on flexible PDMS surfaces. As a result, thiram concentrations and crystal violet (CV) within 10^{-5}M and 10^{-7}M were measured on fish skin and orange peel, which revealed the analytical versatility of SERS substrates**. (Page 2)**

[12] T. Ding, D.O. Sigle, L.O. Herrmann, D. Wolverson, J.J. Baumberg, Nanoimprint lithography of Al nanovoids for deep-UV SERS, ACS applied materials & interfaces, 6 (2014) 17358-17363.
[13] A. Das, U. Pant, C. Cao, R.S. Moirangthem, H.B. Kamble, Fabrication of plasmonic nanopyramidal array as flexible SERS substrate for biosensing application, Nano Research, (2022) 1-9.
[14] M. Gómez, S. Kadkhodazadeh, M. Lazzari, Surface enhanced Raman scattering (SERS) in the visible range on scalable aluminum-coated platforms, Chemical Communications, 54 (2018) 10638-10641.
[15] A. Alyami, A.J. Quinn, D. Iacopino, Flexible and transparent Surface Enhanced Raman Scattering (SERS)-Active Ag NPs/PDMS composites for in-situ detection of food contaminants, Talanta, 201 (2019) 58-64.

In addition, the end of the introduction should introduce the element of novelty of the author's approach. At the same time, in the conclusions section, the authors should highlight the main advantages/differences between the results they obtained and those from similar, already published, works.

Response: Thank you for the comments. There are some novelty points of our study have been mentioned in the end of introduction, "A novel, flexible and reproducible bionic-nanostructure SERS substrate would be demonstrated for the rapid determination of water pollutants, such as heavy metal, toxic molecules, and pesticides". **(Page 3)**

Furthermore, we have highlighted the main advantages in the conclusions section, "Comparing with other references of nano-island substrates (the dragonfly wings [ref. 32]), the beetle wings display unique and natural nanostructure to produce enormous hot-spots for Raman enhancing. Also, the BWS-PDMS based substrate exhibits the flexible, environment-friendly and easier procedure to produce, which makes the chance for the commercialization products". **(Page 9)**

[32] G.C. Shi, M.L. Wang, Y.Y. Zhu, L. Shen, W.L. Ma, Y.H. Wang, R.F. Li, Dragonfly wing decorated by gold nanoislands as flexible and stable substrates for surface-enhanced Raman scattering (SERS), Scientific Reports, 8 (2018) 1-11.

-in the section 3.4, the authors must compare their results, with those reported in other articles (as an example, those they decide to cite in the introduction section, or others), to highlight whether the enhancement factor they obtained are competitive or which could be the reasons of such differences.

Response: Thanks for your suggestion. After comparing those previous reports, the most of references (AuNPs-base) have a higher enhancement factor at 10^5-10^6. Unfortunately, enhancement factors in our system are about 10^4-10^5, because the silver nanoparticle is successive depositing on the bionic nanostructure which will decrease the intensity of SERS signal. However, we use the PDMS replicated method to transfer bionic nanostructure which demonstrates new strategy and possibility to fabricate the SERS substrate.

Although the result of the LOD measurement compared with other similar research [31] was not good enough but this unique 3D bionic structure demonstrates the potential possibility of SERS detection. Also, the PDMS-BWS substrate has more flexibility within the advantages of environment-friendly and easily procedure than the cicada wings. **(Page 8)**

[31] N. Zhao, H. Li, C. Tian, Y. Xie, Z. Feng, Z. Wang, X. Yan, W. Wang, H. Yu, Bioscaffold arrays decorated with Ag nanoparticles as a SERS substrate for direct detection of melamine in infant formula, RSC Advances, 9 (2019) 21771-21776.

Moreover, English has been revised by MDPI English editing center.

We certify that the following article

Flexible PDMS-based SERS Substrates Replicated from Beetle Wings for Water Pollutants Detection

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has undergone English language editing by MDPI. The text has been checked for correct use of grammar and common technical terms, and edited to a level suitable for reporting research in a scholarly journal.
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