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Article reference: JoVE61801

Manuscript title: Treating surfaces with a cold atmospheric pressure plasma using the COST-Jet

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Dear Editor,

Thank you for your email dated July 22nd, 2020. We are pleased to know that our manuscript was rated as potentially acceptable for publication in Journal of Visualized Experiments by all referees, subject to adequate revision and response to the comments raised by the reviewers.

We would like to thank all referees for their time and effort that they have put into assessing the previous version of our manuscript and for their valuable comments.

After carefully considering the comments made in the referees' report, we hereby submit a revised version of our manuscript for your consideration.

Please find attached to this letter a detailed point-by-point response to the referees' reports. In addition, we included a document ("Golda_JoVE_diff.docx") showing the difference between the original manuscript and the current submission.

We look forward to hearing from you regarding our submission and to respond to any further questions and comments you may have.

Yours sincerely,

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Referee Response: Dissipated electrical power and electron density in an RF atmospheric pressure helium plasma jet

This document details changes to manuscript JoVE61801 submitted on August, 19th 2020 following receipt of referees' reports. A detailed point-by-point response for editorial comments and recommendations of referee 1, 2, 3 and 4 is given (shown in blue font). This is followed by a section outlining additional changes made to this updated manuscript. An attachment "Golda_JoVE_diff.docx" enclosed with this submission shows the difference between the original manuscript and the current submission. This has been prepared using Microsoft Word. It is hoped this will clearly detail all amendments to the original article.

Point-by-Point response

Editorial comments:

You will find Editorial comments and Peer-Review comments listed below. Please read this entire email before making edits to your manuscript.

NOTE: Please include a line-by-line response to each of the editorial and reviewer comments in the form of a letter along with the resubmission.

Editorial Comments:

- Please take this opportunity to thoroughly proofread the manuscript to ensure that there are no spelling or grammatical errors.

As suggested, the manuscript has been proofread and checked for errors.

- Please list a minimum of 6 keywords/phrases.

The keywords are listed in the manuscript lines 26-27: Atmospheric pressure plasma, COST-Jet, plasma medicine, plasma treatment, handling protocol, liquid, surface treatment, CAP.

- **Textual Overlap:** Significant portions show significant overlap with previously published work. Please re-write lines 50-52, 56-58 to avoid this overlap.

Lines 50-52, 56-58 have been re-written to avoid overlap with previously published work.

- **Introduction:** Remove the numbered list.

Amended as suggested. The numbered list has been replaced by a key point list.

- **Protocol Language:** Please ensure that all text in the protocol section is written in the imperative voice/tense as if you are telling someone how to do the technique (i.e. "Do this", "Measure that" etc.) Any text that cannot be written in the imperative tense may be added as a "Note", however, notes should be used sparingly and actions should be described in the imperative tense wherever possible.

1) Some examples NOT in the imperative: 1.1, 1.2, 1.3, 1.4, 1.5–1.7, etc.

Text in the protocol section has been changed to imperative tense.

2) Split up long steps (e.g, 2.4, 2.6, etc).

Long steps have been split up.

- **Protocol Detail:** Please note that your protocol will be used to generate the script for the video, and must contain everything that you would like shown in the video. **Please ensure that all specific details (e.g. button clicks for software actions, numerical values for settings, etc) have been added to your protocol steps.** There should be enough detail in each step to supplement the actions seen in the video so that viewers can easily replicate the protocol.

More details have been added to the steps, e.g. button clicks for software actions.

- **Protocol Numbering:** Please add a one-line space after each protocol step.

Amended as suggested.

- **Protocol Highlight:** After you have made all of the recommended changes to your protocol (listed above), please re-evaluate the length of your protocol section. Please highlight ~2.5 pages or less of text (which includes headings and spaces) in yellow, to identify which steps should be visualized to tell the most cohesive story of your protocol steps.

- 1) The highlighting must include all relevant details that are required to perform the step. For example, if step 2.5 is highlighted for filming and the details of how to perform the step are given in steps 2.5.1 and 2.5.2, then the sub-steps where the details are provided must be included in the highlighting.

- 2) The highlighted steps should form a cohesive narrative, that is, there must be a logical flow from one highlighted step to the next.

- 3) Please highlight complete sentences (not parts of sentences). Include sub-headings and spaces when calculating the final highlighted length.

- 4) Notes cannot be filmed and should be excluded from highlighting.

We highlighted the steps of the protocol to be visualized in yellow.

- **Discussion:** JoVE articles are focused on the methods and the protocol, thus the discussion should be similarly focused. Please ensure that the discussion covers the following in detail and in paragraph form (3-6 paragraphs): 1) modifications and troubleshooting, 2) limitations of the technique, 3) significance with respect to existing methods, 4) future applications and 5) critical steps within the protocol.

We addressed all of the above-mentioned categories in the discussion. Due to the complex nature of the plasma chemical treatment, we have sorted the categories by topic. To increase the focus of the discussion, we shortened part of the text. Additionally, we inserted keywords such as „critical steps“ etc. into the discussion to improve orientation.

- **Table of Materials:**

- 1) Please sort in alphabetical order.

Amended as suggested.

- If your figures and tables are original and not published previously or you have already obtained figure permissions, please ignore this comment. If you are re-using figures from a previous publication, you must obtain explicit permission to re-use the figure from the previous publisher (this can be in the form of a letter from an editor or a link to the editorial policies that allows you to re-

publish the figure). Please upload the text of the re-print permission (may be copied and pasted from an email/website) as a Word document to the Editorial Manager site in the "Supplemental files (as requested by JoVE)" section. Please also cite the figure appropriately in the figure legend, i.e. "This figure has been modified from [citation]."

We do not re-use figures that have been published previously.

Reviewer #1:

The Authors present a well written and perfectly described set of actions for using the COST reference plasma jet in the treatment of materials. The paper is certainly fit for JoVE and deserves to be published. Some revisions are anyhow needed to have it published.

- First of all, the title should be changed from "Treating surfaces with a cold atmospheric pressure plasma using the COST-Jet" to something more coherent with the paper content. Results are presented only for a liquid interface and the word "solid" is used only two times besides title and abstract.

We use the word "surface" for both solids and liquids, which also constitute a surface. We changed the heading of section 4 to "(Solid) surface treatment" to clarify that this section is dedicated to the treatment of solids while section 5 is dedicated to the treatment of liquid surfaces. The summary was also amended to make this clearer, now stating: "This protocol is presented to characterize the setup, handling and application of the COST-Jet for treatment of diverse surfaces such as solids and liquids."

- The Authors might give more details on the relative importance for the reproducibility of results of using metal pipes and clean the gas supply lines given as mandatory while they only suggest setting up a controlled atmosphere for the experiments (point 1.8).

We have already addressed both issues in the discussion of the previous manuscript: Firstly, as the purity of the feed gas and thus the tightness of the gas supply system determines the plasma chemistry, this point is relevant of all plasma sources including the COST-Jet. Therefore, it is included in step 1.1 in the protocol: "Set up the gas supply consisting of all-metal gas lines, avoiding any TPFE or similar plastics tubing (Winter²⁴)".

Secondly, the surrounding atmosphere of an atmospheric pressure plasma can potentially influence the reactive species reaching a surface, as well. However, Gorbanev *et al.* have shown that for the COST-Jet this is not the case (Gorbanev, Y. *et al.* Combining experimental and modelling approaches to study the sources of reactive species induced in water by the COST RF plasma jet. *Physical chemistry chemical physics: PCCP*. **20** (4), 2797–2808, 10.1039/C7CP07616A (2018).). Therefore, we only suggest setting up a controlled atmosphere for the COST-Jet.

- The Authors should consider the opportunity of giving more information on the details of figure 4. They should explain if the meniscus is affecting the whole surface of the liquid substrate and why there's no evident dimple under the jet; I might see a dimple, but the Authors do not put that into evidence. Is the meniscus due to the geometry of the experimental area and to the liquid characteristics? Or is it given by the local fluidynamics of the experiment? It would be interesting to understand the reasons for the versus of the flow as shown by the arrows added to the raw picture. Also if it's correct that the jet axis doesn't seem to be centered in the middle of the liquid substrate area. It's particularly strange that the Authors did not mention some recent works where jets (other

than COST reference) impinging on liquid and solid targets are considered and the influence on results of the substrate properties:

A. Stancampiano, E. Simoncelli, M. Boselli, V. Colombo, M. Gherardi, Experimental investigation on the interaction of a nanopulsed plasma jet with a liquid target, Plasma Sources Science and Technology (2018) DOI:10.1088/1361-6595/aae9d0

E. Simoncelli, A. Stancampiano, M. Boselli, M. Gherardi, V. Colombo, Experimental investigation on the influence of target physical properties on an impinging plasma jet, Plasma, Special Issue "Low Temperature Plasma Jets: Physics, Diagnostics and Applications" DOI: 10.3390/plasma2030029

Thanks for pointing out that Figure 4 is not as explicit as we thought it would be. For clarity, we added some more schematic lines to address this issue. The liquid surface on the picture is defined by the blue line showing a tiny dimple directly below the COST-Jet's effluent. The visible edge above this line is created by the capillary action of the liquid inside of the cuvette. We conducted some fluid simulations to understand the movement of the liquid dependent on the impinging gas flow. The direction of the vortices is defined by the surface friction between the liquid and the effluent gas flow streaming above the liquid surface. We would really like to go into more detail regarding the details. However, we were asked to keep the description short here by the editorial comments. There will be a follow-up publication on this topic concentrating on the liquid treatment.

To underline the symmetry of the picture and the centered gas flow, we also added schematic lines to emphasize the COST-Jet's discharge channel contours in the picture.

We added the references to the introduction of the publication.

- Can the Authors comment on any evaporation of the liquid during the experiments?

Evaporation is an issue when treating liquids using cold atmospheric pressure jet sources. In particular for long treatments times and small liquid volumes, the effect on the concentration of species should be considered. Additionally, evaporating liquids might influence the plasma chemistry. In our experiments, we observed evaporation rates in the range of 0,03 ml/min.

- The Authors should describe the experimental configuration of Figure 5: Schlieren images of the COST-Jet with and without applied voltage for two different gas flow rates. Nowhere in the text the reader can understand which is the substrate for those images.

- Also, no information is given on the type of Schlieren imaging setup used by the Authors, even though Schlieren imaging is one of the paper keywords.

- To give the reader a better understanding of the Schlieren imaging techniques used to investigate the interaction with surfaces the Authors might reference the recent "review" paper:

E. Traldi, M. Boselli, E. Simoncelli, A. Stancampiano, M. Gherardi, V. Colombo, G. S. Settles, Schlieren imaging: a powerful tool for atmospheric plasma diagnostic, EPJ Techniques and Instrumentation: Thematic Series on Novel Plasma Diagnostics (2018), DOI:10.1140/epjti/s40485-018-0045-1

The Schlieren images were taken using a single mirror inline arrangement. As Schlieren imaging is not the main topic in this publication, we did not include a detailed description of the setup in the text but added a reference that shows a schematic and a description of the setup. Additionally we added the following sentence to the text: "They show how the horizontally aligned COST-Jet effluent hits a flat glass substrate." Additionally, we removed "Schlieren imaging" from the keywords.

- Why it is true that some of the methods proposed in the paper might be usefully in common use

also for other plasma jet sources, to avoid what might be read as an inflated claim ("It is a stable, reproducible plasma source exhibiting a unique remote character amongst the plethora of different plasma jet designs"), the Authors should also say something on the possible limits of this source, if existent, in really treating solids and liquids for materials of biomedical purposes besides laboratory experiments.

We added a sentence in the introduction to underline the scientific purpose of the COST-Jet: "The COST-Jet is a reliable plasma source that was developed for scientific reference purposes rather than for industrial or medical use".

- The text of reference 15 must be corrected.

Thanks for pointing this out, we corrected the typo.

Reviewer #2:

Manuscript Summary:

The manuscript is written smoothly. Although there is not nay novelty in the manuscript, the authors have tried to explain the protocols to perform reliable and reproducible surface treatments using the COST-Jet.

Major Concerns:

There are few comments that needs to be addressed. This has been suggested in the manuscript. E.g, how did you calculate the power? The phase angle has not been mentioned. Can you please include the formula used for the estimation of power? as there are many formula for estimating the power?

For power calculation, we used the "COST power monitor" software as described in the manuscript it is based on the formula $P = U * I * \cos(\varphi)$ as described in previous publications (J Golda *et al* 2016 *J. Phys. D: Appl. Phys.* **49** 084003, J Golda *et al* 2019 *Plasma Sources Sci. Technol.* **28** 095023). We added these references to the protocol.

It will be better if you could calculate the variation of electron density with the change in voltage, flow-rate.

We agree that this is an important and interesting measurement. Therefore, we did this in a previous publication (J Golda *et al* 2019 *Plasma Sources Sci. Technol.* **28** 095023).

Is the 50Ω current measuring resistor part of experimental setup, or is it used only during the analyses of plasma? If latter, does the resistor influence the plasma properties?

The author has should explain their apparatus/ experimental setup in detail.

The experimental setup is described in detail in previous publications (J Golda *et al* 2016 *J. Phys. D: Appl. Phys.* **49** 084003, J Golda *et al* 2019 *Plasma Sources Sci. Technol.* **28** 095023). As the 50 Ω resistors are part of the oscilloscope, it is not explicitly drawn in the schematics. The resistors are always used when measuring current or voltage. They do not influence the plasma properties as they are only part of the measuring circuit for electrical diagnostics. As advised by the editors of JoVE, we kept the description of the setup as concise as possible and instead added references to previous publications (e.g. J Golda *et al* 2019 *Plasma Sources Sci. Technol.* **28** 095023) instead.

Minor Concerns:

There are minor grammatical mistakes in the manuscript that needs to be addressed. Further, there are repetition of the sentences in the 2.8 and 4.3. Make it short and sweet

We proof-read the manuscript once again and hope to have found all of the grammatical mistakes and typos. The repetitions are due to the protocol that requires a strict description of the required actions.

Reviewer #3:

Manuscript Number: JoVE61801

The manuscript entitled: Treating surfaces with a cold atmospheric pressure plasma using the COST-Jet has been revised.

My comments; It is an interesting and valuable piece of work and worth the publishing after considering the following comments:

- There is no sufficient statement in the introduction to highlight the necessity and importance of the present work.

Due to the the complex conditions in biomedical plasma treatments, standardized sources and protocols might increase the reproducibility of scientific results. Therefore, we decided to make a detailed description available of how to apply the COST-Jet to surfaces. This is underlined in the introduction using the sentence “However, the scientific results often suffer from reliability and reproducibility problems” which cites two articles in *Nature* depicting the challenge. To make this even more clear, we added another sentence in the introduction: “Compliance with the protocol presented here should ensure the reproducibility and comparability of the measurements.”

Reviewer #4:

Manuscript Summary:

Authors demonstrate a protocol for the COST-Jet, 1 mm plasma exit for surface/liquid treatment.

Major Concerns:

Instead of the voltage to power plot (figure 2), should authors provide reactive species to power plot as a representative result to fulfil the surface/liquid treatment (figure3,4). The COST-Jet aims to stand for He-O jet plasma source, then the O-atom dose that reach the surface(s) might mostly be informed in the protocol.

We also think that the oxygen density in dependence of the applied power and target distance is an important and interesting measurement. Therefore, these measurements have already been performed and published in previous papers (e.g. Gert Willems *et al* 2019 *New J. Phys.* **21** 059501 or Frederik Riedel *et al* 2020 *Plasma Sources Sci. Technol.* in press <https://doi.org/10.1088/1361-6595/abad0>) which are also referenced in the text. We added a sentence to the representative results section: “More detailed measurements of reactive species (e.g. atomic oxygen and ozone), temperature and power as well as bactericidal activity measurements have been performed by Riedel *et al.*²³.”.

Additional corrections and amendments

We additionally exchanged single words and corrected misspellings for better readability. For details, please see attached ‘Golda_JoVE_diff.docx’.