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Interactive comment on "Open-source, 3D-printed, high-pressure (50 bar) liquid-nitrogen-cooled *para*-hydrogen generator" *by* Frowin Ellermann et al.

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Dear James Eills,

We very appreciate your contribution to our manuscript. We implemented all of your comments. Hereby, we want to provide a point-by-point response to your comments.

Your comment: Title: "3D-printed" should be removed from the title or clarified as "partially 3D printed"(or similar), as this is misleading in its current form.

Answer: We agree. And to avoid this confusion modified the title as follows:
New title: "Open-source, partially 3D-printed, high-pressure (50 bar) liquid-nitrogen-cooled parahydrogen generator"

Your comment: Line 40: The authors might choose to also mention long-lived spin isomers in methylrotors, e.g. doi: 10.1021/ja410432f
Answer: Thanks for pointing out this paper! We added it to the list.
Old text: "Hydrogen is not the only compound that has stable or long-lived spin isomers at room temperature (rt) there are many examples: deuterium (Knopp et al., 2003), water (Mammoli et al., 2015; Meier et al., 2015), ethylene (Zhivonitko et al., 2013), and even naphthalene derivative (Stevanato et al., 2015)."
New text: "Hydrogen is not the only compound that has stable or long-lived spinisomers at room temperature (rt) there are many examples: deuterium (Knopp et al., 2003), water (Kravchuk et al., 2011; Vermette et al., 2019), ethylene (Zhivonitko et al., 2013) and methyl groups (Meier et al., 2013)."

Your comment: Line 113: Can the authors make any comment about how well the cotton wool works in preventing iron oxide flow through the system, or state how much was used for effective filtering?
Answer: To address this in the text we modified and added one section in the text. Now it is as follows
New text: "On both ends of the copper coil, cotton wool was pressed to keep the catalyst in place to protect the rest of the system from contaminations. The compressed wool insets have a length of 20 mm. Wool as a particulate filter was used before in another PHG (Du et al., 2020). During the six months of weekly use of our generator, there was no sign of a moving catalyst."

Your comment: Line 141: It would be helpful to briefly state how inductive and static

spark charges are avoided.
Answer: That's a good point! In contrast to fluids, gas does not induce charges in pipes. We modified the safety concept and added a sentence to the discussion.
Old text: "iv. Avoidance of inductive and static spark charges"
New text: "iv. Avoidance of inductive and static spark charges in the gas lines (due to conductive and groundable pipe material)"
And in discussions: "The risk for static and inductive spark charges in the gas line is low (Department of Labour of New Zealand, 1990). Nevertheless, the gas pipes can be grounded to prevent electrical charges on the parts which are in contact with H_2 gas."

Your comment: Line 265: I'm a little bit confused about the reporting of p_{out}, doesn't this value vary in time? Does p_{in} describe the gas pressure in the catalyst region? Isn't this the only relevant pressure when considering conversion as a function of flow rate?
Answer: We agree on this point. p_{out} is varying during filling and now we defined p_{target} that is final p_{out} value. We changed the whole manuscript accordingly.

New text:
Definition:
We refrained from including a flow meter in the setup to keep it simple and robust. Instead, we used the time $_{tp,V}$ needed to fill a cylinder of a given volume V_0 to a given pressure p_{target}...
And example:
"For the latter, the para-enrichment was found to be constant up to a flow rate of f_r = 2 SLM (for p_{in} = 20 bar, p_{target} = 10 bar)."

Your comment: Lines 343-344: But the quantification is performed on a high-field

NMR system. Better to say "We have shown that routine pH2 quality control can be performed with a low-cost benchtop NMR system", or similar.
Answer: We agree and modified the sentence.
Old text: "Both values are suited for routine pH_2 quality control without a need for an expensive high-field NMR system."
New text: "These results indicate, that the routine pH_2 quality control can be performed with a low-cost 1 T benchtop NMR spectrometer."

Your comment: Lines 347-351: I don't understand the value "points loss of fpH2 per week". A relaxation time is given to describe the exponential process and this should be sufficient to understand thefpH2change each week. Unless I've misunderstood, I suggest simply removing this.
Answer: This value is another way to report the lifetime, but it doesn't transfer additional information. We completely removed this paragraph because it seems to be confusing, also to other referees.

Your comment: Line 348: "With 120 days of lifetime, Hövener et al. reported even longer values(2013)." The phrasing of this sentence is a little strange.
Answer: We agree and rephrased the paragraph.
Old text: "Feng et al. reported a lifetime in aluminium tanks of (63.7 $\pm$ 8.3) days and about 2 % points loss of f_{pH_2} per week (2012). With 120 days of lifetime, Hövener et al. reported even longer values (2013).
New text: "The relaxation time constant in aluminium tanks was found to be (63.7 $\pm$ 8.3) days by Feng et al. (2012) and 120 days by Hövener et al., respectively (2013)."

Your comment: Line 349: It would be helpful to explain why vacuuming the cylinder should lead to a longer pH2lifetime (presumably due to removal of paramagnetic oxygen molecules).

Answer: We removed the part related to vacuuming since we did not perform a cleaning procedure with the storage bottles. However, the production protocol features a flushing routine which removes potential air contamination in the cylinder.
Old text: "Note, that we did not vacuum our cylinder that can increase the lifetime."
New text: "Note, that we did not perform any dedicated cleaning procedure for the pH_2 storage bottle."

Your comment: Figure 7: It would be informative to include an additional plot showing para-enrichment level as a function of pressure (Pin) for a 2 SLM flow rate. This is a suggestion and I do not insist on this for publication.
Answer: We did not observe any pressure dependency for the p_{in} values we investigated (12 bar, 20 bar, 35 bar, 50 bar) and now it is mentioned in the text:
New text: "We also investigated f_{pH_2} as a function of the p_{in} pressure at a fixed flow rate: a batch was prepared for p_{in} equals 12 bar, 20 bar, 35 bar and 50 bar, p_{target} = 10 bar and a flow rate of 0.9 SLM. No pressure dependency could be observed. The obtained average of f_{pH_2} is (52.4 $\pm$ 0.8) %."

Your comment: In another interactive comment, Prof. Igor Koptyug has provided two references to other parahydrogen generators described in the literature. I agree that this closely-related work should be cited.
Answer: The papers were published after we finished our manuscript. Nevertheless, they provide valuable information to our paper and we included both He-based PHGs into our overview table.

Thank you very much again for your extensive review, and we hope, that we could address all your comments appropriately.

With kind regards,
Frowin Ellermann and Jan-Bernd Hövener

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