nature portfolio

Peer Review File

Record stack durability in industrial solid oxide steam electrolysis through operational control

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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 share 25 kh of operation of an SOEC full stack using a novel ACDC operating strategy. The durability data is completed with EIS and post mortem analyses. These are outstanding and ground breaking results, and I want to congratulate everybody that took part in it, from the stack design and manufacturing teams to the SEM operators, and everybody in between. Reaching long operating times at stack level is hard work, and you absolutely shattered the previous record!

Below are some comments.

51: At stack level, voltage increase would be accompanied by a temperature increase, and decreased efficiency (or higher electrical cost). Conveniently, stack temperature can "be increased" to stabilize both i and U. I would argue that the operational strategy you are describing here is hardly typical anymore, especially since stacks are getting more powerful.

53: The term "industrially relevant lifetimes" appears multiple times throughout the paper and should be defined. Are we talking about 20, 40, 80, 350+ kh ?

56/57 This statement appears to be a repeat of line 53/54.

57: continuous degradation at 0.01 %/kh would have a limited impact on LCOH, yet high degradation / short lifetime certainly would. The degradation being continuous does not seem to be the core of the limitation.

73: this statement needs references.

83: If thermal balancing is the objective, then potentio-galvanostatic operation at the thermoneutral voltage seems the far easier solution compared to rapidly alternating the current, a functionality that may complexify power electronics. It seems to me that alternating the current is wanted to suppress electrochemical gradients, which then leads to choosing endothermic operation during the SOEC operating window to balance global heat generation. If so, maybe the text could be clarified a bit.

107: Though the statement is vague enough, it suggests stacks generally fail due to "normal" degradation. I would argue that we have yet to see a single report of SOEC stack test stopped due to low performance (eg potentiostatic SOE stack where current is now near 0 due to degradation). Most, if not all stacks (outside of yours!) fail to a loss of tightness (refs 36 and 38 did, and the model used by ref 41 extrapolates stack lifetimes in the 60+kh range from single cell test data over 1-4kh). Whether that loss of tightness is due to progressive degradation, or initial manufacturing defects remains to be seen (and maybe that distinction is not even warranted).

129: Could the authors explicit how the conditions were chosen ? Is it the cycle duty that was chosen to reach overall thermal gradient at 750°C ? What the duty set to about 90% and currents optimized?

Supplementary note 2: Could the authors explicit how the stack temperature was defined and measured? I'm assuming the stack was gas-heated, but this should be specified. Was the stack inside a furnace or a hotbox without temperature

actuators?

Fig S3 could likely be improved by improving its quality (maybe it's the reviewers version ?), by increasing Ymin, and normalizing per RU for easier comparison. Similarly, there is really no real need to have the Yaxis of S2a go from 0 to 1000°C, when a quarter of that scale would be sufficient. Outside of that, all visuals are great !

130-134: I'm not sure what point you are making here. Is it that standard DC operation typically leads to measurable (non-nil) degradation? Didn't DTU show that cleaning impurities cancelled degradation (albeit over 500h), even under DC operation (10.1149/1.3455882) ?

136: could the authors elaboration on that conditioning period? 2,700h seems particularly long to reach stable/linear behavior. 500-1000 h seems more typical for CSC.

143 & Table S2: Didn't AC/DC operation during phases 2 & 3 lead to monotonic increase of voltage and degradation rates in the range you use to contrast the rest of the test in AC/DC ? Could the authors expand on this ? Are the extremely low DR obtained at 750°C the consequence of the operating strategy, or the intrinsic extreme stability of the stack/cells ? Phase 5 (no degradation at 750°C and DC mode over 600h) would suggest the latter and contradict line 142.

147: Similarly, I'm not sure you fully show that the outstanding results are a consequence of the operating mode, since DC operation in phase 5 did not lead to an increase in voltage.

153-155: Unless I'm missing something, if standard thermoneutral operation, which effectively both stabilizes voltage (1.29 V/RU) and eliminates thermal gradients, does lead to monotonic degradation, then these cannot be the activators of dominant degradation pathways that AC/DC mode suppresses. Can the authors comment ?

196: I would say that the electrochemical response was indeed stable during AC/DC operation, but based on the results shared, I'm not sure the causality is clearly shown.

222: The wide range of degradation rates found in the literature would suggest that materials, microstructures, manufacturing process, operating parameters and quality of utilities (likely among others) all play a role in the stability (or lack thereof) of the stacks during standard DC operation. While it is very much warranted here, the comparison would be ideal if SolydEra had made no improvement to their cells over the past decade, and if utility quality was same during both tests. Both are likely not true.

287: Before jumping into economics, it would be quite interesting to discuss the limitations of the ACDC strategy. For example, the strategy decreases the maximum SC achievable, a parameter with high impact on efficiency (even assuming free/low cost vaporization). In your case, the 76% SOEC SC goes down to 63% overall. Describing the non-achievable window of SOEC operation would be a great addition, as well as the (module) efficiency cost. Similarly, how could the control parameter of the strategy evolve with degradation (even if low)? Voltage increase over time means that the stack will be less endothermic in SOEC, and becomes exothermic overall. Conversely, could this strategy help with heat generation in SOFC? And finally, what are the implications on large systems power electronics specifications? Is it limited, with current solutions being (somewhat) compatible? Should a new generation of solutions be developed?

299: Could the authors expand on their lifetime definition? Is it a maximum voltage / minimum acceptable conversion efficiency? If so, as long as the stack and its tightness are functional, what is stopping the operator to increase the temperature and expand the lifetime? Also, putting forward a 350+ kh projected lifetime likely calls for a sentence or 2 about aging of the tightness solution. The results here do not cover the stability of the glass ceramic matrix, and the vast majority of long-term tests end due to tightness failure.

327-363, 347 in particular, and overall:
This works shows early degradation in ACDC mode, and no degradation throughout a later short DC operating window. I don't believe the latter result is properly discussed, given its massive implication. Regarding the statement of 347, this can only truly be known by switching a DC operated stack that degrades significantly to ACDC operation and showing some form of cancelation. Also, using a SolydEra stack for your demonstration does not really work in your favor, since their cells/stacks do show great stability even when DC operated. Also, 347 can only be stated if/when your results are compared to an equivalent DC test, which you do not have. Selected use of conditional might be warranted. The outstanding and impactful nature of the work done shouldn't be lessened in any way if/when DC data of SE stacks showing low degradation is published.

Reviewer #2

(Remarks to the Author)
Dear authors,

thank you very much for this interesting and impressive paper about SOEC long-term operation. I have only some minor comments/questions, polease refer to the pdf attached.

Reviewer #3

(Remarks to the Author)
The manuscript describes a novel operating AC:DC strategy for minimizing degradation of SOEL stacks.
The manuscript has a very high quality and is well written.
The methodology and results are scientific, well discussed and sound.
The established literature in this topic is up-to date and good referenced.
The conclusions are underlined trough different evaluation methods (long-term operation, EIS and post-mortem analysis)
The results are important and significant for the SOEL community.

Minor review recommendations are:
1) Format: use fixed blanks between numbers and units
2) Format: The fond size of figures S14a and S17a is very small. Please increase.
3) The structure of the manuscript is separated into several sections (main text, methods, figure captions, and supplement), which are not logically ordered. Hence, it is very difficult to read the manuscript. It is recommended for the final manuscript to merge the information in logical sections.
4)The degradation of the stack is mostly only given as relative value. Please also add the absolute degradation per cell in mv/kh.
5) Please describe the thermocouple positions and the calculation of the nominal average stack temperature more clearly.
6) The stack performance at the beginning of operation is not reported in the manuscript. Please give the important initial stack data: electrolysis power, steam conversion rate, electrical stack efficiency and produced flow rate of H2.
7) Usually the degradation at the beginning of operation is higher compared to extended operation periods. Please discuss the impact of time on the low degradation in context to the impact of AC:DC method.
8) Please shortly specify/discuss the 5 different electrochemical processes identified in the DRT spectra.
9) According to Fig. 2d the contribution of R4 is in the same range as Rs and even higher compared to R2. Please comment the contribution of R4.
10)The cost of stack is given in €/m2, which is quite unusual. Should it not be €/kW or €/MW? Please comment. (see also comment in Supplement Table S8)
11)Add doi numbers of references (if available).

Both reviewed documents (manuscript and supplement) with all changes (highlighted) and all comments are attached.

Reviewer #4

(Remarks to the Author)
This is an interesting work, which advances the SOFC development. I suggest the publication of this work after considering the following comments.
1. There is a transient behavior before the stabilization (the first 3000 hours). What happens during this period? A more detailed analysis during this period may help the readers' understanding.
2. The AC:DC waveform uses 30 Hz. Why this frequency is used? Some discussion and justification is needed.
3. An averaged DRT analysis is given across all eight cell groups, which is interesting but also risks masking heterogeneity. I suggest the authors also show the DRT of each group.
4. The post-mortem analysis is mostly qualitative. Quantitative analysis, such as the particle size of nickel, porosity…., is suggested to be provided.
5. Although a short-stack DC test is compared, no long-duration DC control test on the same stack design is compared, making the direct comparison of the AC:DC control somehow weak. I can understand additional test on this issue will take too much time. If such a test is not feasible to do, some explanation is preferred.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
I want to thanks the authors for the thoughtful answers to the reviewers' comments, and the insightful discussions. I have no notes.

Congratulations again for the achievement ! I'm looking forward to the study being published.

Reviewer #3

(Remarks to the Author)
Dear authors of the manuscript,

Thank you for adressing all my reviewer comments.
The manuscript can be published as it is (without further changes).

Reviewer #4

(Remarks to the Author)
The revision is good, and I have no further comments. My suggestion is to accept this paper for publication.

REVIEWER COMMENTS
Reviewer #1 (Remarks to the Author)

The authors share 25 kh of operation of an SOEC full stack using a novel ACDC operating strategy. The durability data is completed with EIS and post mortem analyses. These are outstanding and ground breaking results, and I want to congratulate everybody that took part in it, from the stack design and manufacturing teams to the SEM operators, and everybody in between. Reaching long operating times at stack level is hard work, and you absolutely shattered the previous record!

Authors: *We sincerely thank the reviewer for the very positive assessment of our work and for the generous recognition of the efforts behind this long-term stack experiment. We greatly appreciate the reviewer's acknowledgment of both the technical challenge and the collaborative nature of achieving stable 25,000 h SOEC stack operation. We are encouraged that the reviewer considers the results outstanding and impactful for the SOEC field.*

Below are some comments.

51 (51-63 in the updated draft): At stack level, voltage increase would be accompanied by a temperature increase, and decreased efficiency (or higher electrical cost). Conveniently, stack temperature can "be increased" to stabilize both i and U. I would argue that the operational strategy you are describing here is hardly typical anymore, especially since stacks are getting more powerful.

Authors: *We thank the reviewer for this important comment. We agree that, at the stack level, temperature regulation can partially compensate for the voltage increase and help maintain stable voltage operation under conventional DC galvanostatic electrolysis. Our intention was not to imply that voltage stabilization is achieved through thermal adjustment, nor that such operation remains representative of current industrial practice for increasingly powerful stacks. To clarify this point, we have revised the text to better distinguish between conventional thermal management strategies and the present AC:DC operational approach. We now emphasize that the AC:DC proposed strategy is not based on increasing stack temperature, but on dynamically modulating electrochemical polarization and heat generation through periodic polarity inversion. We have also softened the wording regarding "typical" stack operation and clarified the associated efficiency and operational implications in the revised manuscript.*

53 (51-63 in the updated draft): The term "industrially relevant lifetimes" appears multiple times throughout the paper and should be defined. Are we talking about 20, 40, 80, 350+ kh ?

Authors: *We thank the reviewer for this important clarification. We agree that the term "industrially relevant lifetimes" was insufficiently defined. In the revised version, we now specify the relevant operational ranges more explicitly and avoid using the term in a generic way. In particular, we clarify that current industrial expectations for SOEC stack operation are*

generally in the range of several tens of thousands of hours, with practical deployment targets often exceeding 40,000–80,000 h depending on system design, replacement strategy, and application.

We have also revised the techno-economic analysis in the Methods section to clarify how stack lifetime is treated in the LCOH calculation. Specifically, we now state that stack lifetime was defined by a thermal end-of-life criterion, in which voltage degradation increases electrical energy input and heat generation during galvanostatic operation, progressively shifting the stack's thermal balance. In the analysis, the stack was considered to reach the end of life when the calculated outlet temperature reached 800 °C, which was taken as the maximum allowable operating temperature.

We further clarify that the longer projected lifetimes discussed in the techno-economic analysis correspond to model-based extrapolations of electrochemical degradation trends and should not be interpreted as experimentally demonstrated full-system lifetimes.

56/57 This statement appears to be a repeat of line 53/54.

Authors: *We thank the reviewer for this note. We fixed the text to avoid this repetition.*

57 (51-63 in the updated draft): continuous degradation at 0.01 %/kh would have a limited impact on LCOH, yet high degradation / short lifetime certainly would. The degradation being continuous does not seem to be the core of the limitation.

Authors: *We agree with the reviewer. The critical limitation is not the mere presence of continuous degradation, but degradation rates high enough to reduce efficiency, shorten usable stack lifetime, and increase LCOH and stack replacement costs. We have revised the text accordingly. The manuscript now distinguishes practically tolerable voltage drift from degradation that materially affects LCOH and system lifetime. We also removed wording that could imply that any continuous degradation is intrinsically limiting.*

73 (77-79 in the updated draft): this statement needs references.

Authors: *We thank the reviewer for this note. We fixed the text by adding the references below.*

- Taubmann, J. et al. Tracking degradation in solid oxide cells by a coupled time and frequency domain multi-physics model: A case study on nickel migration in electrolysis mode. J. Power Sources 659, (2025). https://doi.org/10.1016/j.jpowsour.2025.238388
- Chatzichristodoulou, C., Chen, M., Hendriksen, P. V., Jacobsen, T. & Mogensen, M. B. Understanding degradation of solid oxide electrolysis cells through modeling of electrochemical potential profiles. Electrochim. Acta 189, 265–282 (2016). https://doi.org/10.1016/j.electacta.2015.12.067

83(85-89 in the updated draft): If thermal balancing is the objective, then potentio-galvanostatic operation at the thermoneutral voltage seems the far easier solution compared to rapidly alternating the current, a functionality that may complexify power electronics. It seems to me that alternating the current is wanted to suppress electrochemical gradients, which then leads to choosing endothermic operation during the SOEC operating window to balance global heat generation. If so, maybe the text could be clarified a bit.

Authors: *We thank the reviewer for this insightful comment. We agree that, if the sole objective were thermal balancing, potentio-galvanostatic operation near the thermoneutral voltage would indeed be a simpler strategy than rapidly alternating the current. However, the purpose of the AC:DC strategy is broader than thermal management alone.*
More specifically, polarity inversion is primarily intended to periodically relax sustained electrochemical polarization and suppress the buildup of electrochemical gradients and overpotential, while the alternation between SOEC and SOFC operation also contributes to balancing the overall heat generation of the system. We updated the text to emphasize that AC:DC operation should not be interpreted solely as a thermal-management strategy, but rather as a dynamic polarization-management strategy in which both electrochemical relaxation and thermal balancing are regulated.
This interpretation is also consistent with previous economic analysis, which showed that, under galvanostatic operation, it is beneficial to start from an endothermic operating point (https://doi.org/10.1016/j.ijhydene.2025.01.028). This provides thermal headroom, since voltage degradation gradually increases the electrical energy input and, consequently, the heat generation over time. In this context, AC:DC operation may be particularly relevant when endothermic SOEC operation is already preferred from a system-design perspective.

107 (115-120 in the updated draft): Though the statement is vague enough, it suggests stacks generally fail due to "normal" degradation. I would argue that we have yet to see a single report of SOEC stack test stopped due to low performance (e.g. potentiostatic SOE stack where current is now near 0 due to degradation). Most, if not all stacks (outside of yours!) fail to a loss of tightness (refs 36 and 38 did, and the model used by ref 41 extrapolates stack lifetimes in the 60+kh range from single cell test data over 1-4kh). Whether that loss of tightness is due to progressive degradation, or initial manufacturing defects remains to be seen (and maybe that distinction is not even warranted).

Authors: *We thank the reviewer for this important clarification. We agree that the original wording was overly broad and could be interpreted as implying that SOEC stacks generally reach end-of-life solely due to gradual electrochemical performance degradation. As correctly pointed out by the reviewer, long-term SOEC stack tests reported in the literature are terminated due to loss of tightness or other system-level failure modes rather than because the stack can no longer sustain practical electrolysis performance. Furthermore, it is often difficult to distinguish whether such failures originate from progressive degradation mechanisms, manufacturing defects, or a combination of both.*
To address this concern, we have revised the text to avoid implying that electrochemical degradation is the only lifetime-limiting factor in SOEC stacks. The revised manuscript now

states that SOEC stack lifetime can be limited by both progressive electrochemical degradation and system-level failure mechanisms, such as loss of tightness, which together impact long-term operation and economic viability. We also clarify that the present work focuses primarily on electrochemical and microstructural indicators of stack stability, including voltage evolution, impedance response, and post-mortem analysis, rather than on establishing the ultimate sealing or mechanical lifetime of the stack.

129: Could the authors explicit how the conditions were chosen ? Is it the cycle duty that was chosen to reach the overall thermal gradient at 750°C ? What the duty set to about 90% and currents optimized?

Authors: *We thank the reviewer for this important question. In the revised manuscript, we have clarified that the AC:DC operating conditions were selected based on three main criteria:(i) maintaining the desired average current density,(ii) achieving an overall near-zero thermal imbalance across the stack, and (iii) keeping the minimum instantaneous cell voltage during the reverse pulse at approximately 0.72 V per cell.*
In practice, the current amplitudes and duty cycle were adjusted experimentally to simultaneously satisfy these constraints. Thus, the waveform parameters were not determined by a single optimization variable alone, but rather by co-adjustment to achieve stable stack operation while preserving the desired thermal behavior and electrochemical operating window.
*For the operating conditions reported here, this resulted in duty-cycle values of approximately 87–90%, allowing an average electrolysis current density of -0.5 A*cm^{-2}. After approximately 2,700 h, only minimal further adjustment was required, and the operation converged to a duty cycle of about 90% under constant conditions.*
The AC:DC frequency was fixed at 30 Hz and not optimized in the present study. This choice is consistent with earlier AC:DC SOEC work (https://doi.org/10.1016/j.jpowsour.2022.231040) and is also supported by a later study (https://doi.org/10.1016/j.jpowsour.2026.239904). Since the purpose of the present work was to investigate long-term stack durability rather than to optimize waveform parameters, one waveform configuration was selected and then maintained as consistently as possible during the long-term test.
We have clarified this rationale in the revised manuscript and expanded the description of the waveform selection in the Methods and Supplementary Note 2.

Supplementary note 2: Could the authors explicit how the stack temperature was defined and measured? I'm assuming the stack was gas-heated, but this should be specified. Was the stack inside a furnace or a hotbox without temperature actuators?

Authors: *We thank the reviewer for this important request for clarification. We have expanded Supplementary Note 2 to provide a clearer description of how the stack temperature was defined and measured.*

The stack was operated inside an insulated hotbox, not inside a furnace, and there was no active heating of the hotbox during operation beyond the heat generated by the stack itself under AC:DC operation. In particular, the stack was not gas-heated, and no additional electrical heat tracing was applied to the hotbox. The inlet gases were preheated to 750 °C using external heat tracing before entering the stack.

Under these conditions, the reported stack temperature reflects the thermal state of the stack/hotbox assembly. The temperature drop observed during EIS measurements at OCV is attributed to the absence of active hotbox heating under open-circuit conditions; it therefore primarily reflects heat loss from the hotbox when the stack is no longer generating sufficient internal heat.

To further clarify the temperature measurements, we now specify that the thermocouples were positioned in-line along the gas paths, directly at the interfaces between the gas manifolds and the stack. Temperatures were thus recorded at both the fuel-electrode (FE) and air-electrode (AE) inlets and outlets.

We further clarify that the reported nominal temperature difference across the stack was calculated as:

$$\Delta T_{nom} = \frac{\left(T_{FE,out} + T_{AE,out}\right) - \left(T_{FE,in} + T_{AE,in}\right)}{2}$$

This quantity therefore represents the average increase in gas temperature across the stack, based on the measured inlet and outlet temperatures on both the fuel-electrode and air-electrode sides.

Fig S3 could likely be improved by improving its quality (maybe it's the reviewers version ?), by increasing Ymin, and normalizing per RU for easier comparison. Similarly, there is really no real need to have the Yaxis of S2a go from 0 to 1000°C, when a quarter of that scale would be sufficient. Outside of that, all visuals are great !

Authors: *We thank the reviewer for this helpful suggestion and for the positive assessment of the figures. In response, Figs. S2a and S3 have been revised by increasing the image resolution and by adding voltage normalization per repeating unit (RU), as requested. These changes improve readability and facilitate a more direct comparison between cell groups.*

For Fig. S2a, the broader y-axis range was retained intentionally to provide sufficient visual separation between the two temperature profiles. A narrower scale would place the curves too close together, making it harder to distinguish their individual trends and temporal variations. The selected range also allows the use of rounded y-axis values, which improves clarity.

130-134 (140-145 in the updated draft): I'm not sure what point you are making here. Is it that standard DC operation typically leads to measurable (non-nil) degradation? Didn't DTU show that cleaning impurities cancelled degradation (albeit over 500h), even under DC operation (10.1149/1.3455882) ?

Authors: *We thank the reviewer for this important comment and agree that the original wording was not sufficiently precise. Our intention was not to overlook previous studies showing strongly reduced degradation under carefully controlled operating conditions, including the work cited by the reviewer. Indeed, the presence of impurities appears to make a significant contribution to SOEC degradation, and limiting their concentration by filtering the fuel gas can have a clear beneficial impact. However, impurities are not only introduced through the gas feed; they may also originate from the cell and stack materials themselves (https://doi.org/10.1016/S0167-2738(03)00147-4, https://doi.org/10.1016/S0167-2738(02)00269-2, https://doi.org/10.1016/S0167-2738(01)00969-9). During operation, such species can segregate toward electrochemically active regions and block triple-phase boundaries, thereby contributing to performance loss.*
In addition, our previous studies have shown that Ni can migrate away from the electrolyte above a specific cathodic overpotential, even when no detectable impurity accumulation is observed in post-mortem analysis (https://doi.org/10.1149/1945-7111/add298, https://doi.org/10.1016/j.jpowsour.2024.234490). Under sustained unidirectional DC polarization, the synergetic impact of continuous Ni coarsening and Ni migration builds up overpotential (or, in other words, decreasing pO_2 in the TPBs), which can eventually lead to loss of contact between Ni and YSZ. This can manifest as cracks or detachment features in SEM analysis and appears to activate more extensive microstructural degradation of the Ni–YSZ fuel electrode, leading to an accelerated increase in ohmic resistance.
We have therefore revised the text to clarify that measurable degradation is commonly observed during long-term SOEC stack operation under conventional DC electrolysis, but that the degradation trajectory depends strongly on gas purity, material-derived impurities, thermal management, electrochemical history, and the accumulation of cathodic overpotential. In this context, AC:DC operation is proposed to suppress continuous Ni migration and limit the build-up of fuel-electrode overpotential, thereby avoiding the overpotential threshold associated with accelerated ohmic degradation.

136 (147-55 in the updated draft): Could the authors elaboration on that conditioning period? 2,700h seems particularly long to reach stable/linear behavior. 500-1000 h seems more typical for CSC.

Authors: *We thank the reviewer for this important observation. In the revised manuscript, we have clarified that this period should not be interpreted as a conventional short conditioning stage alone, but rather as an investigation/optimization period.*
During this early phase, several operating parameters were varied, including transitions between DC and AC:DC operation and changes in temperature and current density, all of which contributed to the electrochemical evolution observed. A closer inspection of the data suggests that the system may already have been approaching stable behavior after

approximately 2,100 h. However, at around 2,700 h, the operating conditions were fixed and subsequently kept constant. We therefore define the following interval (>22,000 h) as the stabilized operating regime under constant conditions. Importantly, in a full-stack test, the apparent run-in period reflects not only electrochemical conditioning but also equilibration of system-level components and boundary conditions, including preheaters, metallic components, and parameters such as air humidity, temperature, and pressure. For this reason, direct comparisons with typical run-in periods reported for single-cell tests should be made with caution, since those are generally governed much more closely by the cell's intrinsic electrochemical behavior. This distinction has now been discussed in the revised manuscript.

143 & Table S2 (156-161 in the updated draft): Didn't AC/DC operation during phases 2 & 3 lead to monotonic increase of voltage and degradation rates in the range you use to contrast the rest of the test in AC/DC ? Could the authors expand on this ? Are the extremely low DR obtained at 750°C the consequence of the operating strategy, or the intrinsic extreme stability of the stack/cells ? Phase 5 (no degradation at 750°C and DC mode over 600h) would suggest the latter and contradict line 142.

Authors: *We thank the reviewer for this important comment and agree that stack quality is a clear factor. However, across the SOEC literature, DC operation at around 750 °C has generally not, by itself, been sufficient to deliver the level of durability and performance observed here, with signs of Ni migration or YSZ electrolyte degradation being reported. In the present study, we report not only the longest SOEC stack test to date, but also the lowest degradation rate observed at the stack level, with suppressed Ni migration and not observable degradation of the 8YSZ electrolyte. Notably, approximately 23,000 h of the total 25,000 h test duration, corresponding to about 90% of the experiment, were conducted under AC:DC operation.*

We agree that no measurable degradation was observed during the DC period in phase 5. However, this period lasted just 600 h and was followed by extended AC:DC intervals, which appeared to markedly reduce the voltage degradation rate, from 11.5% to 0.6%. We therefore consider it more appropriate to interpret the stable behavior observed during phase 5 as occurring after prolonged conditioning under AC:DC operation, rather than as evidence that DC operation alone would have produced comparable long-term performance. Given the current understanding of SOEC degradation under continuous DC polarization, the beneficial effect already observed during the early AC:DC periods, and the fact that the aim of this study was specifically to examine the impact of AC:DC operation on stack durability beyond 3,000 h, extending the DC mode further than 600 h was considered unnecessarily risky.

The reason is that continuous unidirectional polarization imposes sustained electrochemical, thermal, and mechanical gradients across the stack, thereby increasing the likelihood that a local failure in a single cell could trigger accelerated degradation that would propagate through the stack. In this context, the dynamic relaxation of unidirectional polarization through AC:DC operation may not only mitigate degradation directly but also reduce the likelihood that local unexpected external/internal instabilities develop into broader failures. Another possibility is that the beneficial role of AC:DC is most critical during the early operating

period, for example within the first 1,000–2,000 h, by facilitating the removal of impurities originating from the SOC raw materials until they are completely clean from impurities, and by limiting Ni migration until the Ni matrix, and more generally the SOC microstructure, approaches a more stable thermodynamic state where Ni and potentially other elements cannot move. Under such conditions, overpotential build-up can remain low, well below the threshold for Ni/YSZ detachment, which appears to be associated with the accelerated increase in ohmic resistance. However, this hypothesis cannot be established from the present experiment, because more than 90% of the total operating time was conducted in AC:DC mode.

Importantly, even after apparent stabilization, unexpected external disturbances or internal local failures during DC operation could still initiate a cascade of degradation phenomena. The relaxation of gradients that AC:DC operation imposes may therefore not only suppress degradation but also improve the resilience of the SOEC stack against unexpected failure events.

We have therefore revised the manuscript to avoid implying that the outstanding durability arises solely from the operating mode. Instead, we present AC:DC operation as a plausible and potentially the most important contributor to the stabilization pathway, and possibly also to the resilience of the stack against unexpected internal or external perturbations, while keeping our central claim focused on what is directly demonstrated experimentally: stable operation of a full industrial 70-cell SOEC stack for 25,000 h, with a degradation rate of only 0.05 % kh^{-1} over more than 22,000 h.

147 (156-161 in the updated draft): Similarly, I'm not sure you fully show that the outstanding results are a consequence of the operating mode, since DC operation in phase 5 did not lead to an increase in voltage.

Authors: *We thank the reviewer for this insightful observation. As noted in our response to the previous comment (143), the stable DC interval occurred only after extended operation under AC:DC conditions and therefore represents only a small fraction of the total 25,000 h test duration. We agree that, although our results demonstrate exceptionally stable operation under predominantly AC:DC conditions, they do not prove that AC:DC operation alone is exclusively responsible for the observed durability, particularly since an identical long-term test under pure DC mode is not reported in our study.*

Nevertheless, additional support for our reasoning in the previous comment regarding the beneficial effect of AC:DC operation is provided by recent results reported by Ouwelthes et al., "Steady-state operation of 70-cell SOEC stack – pushing the boundaries of long-term operation at low degradation rates" (EFCF 2026, A1201; no DOI currently available). For completeness, we attach the main figure from that work here but not in the main text.

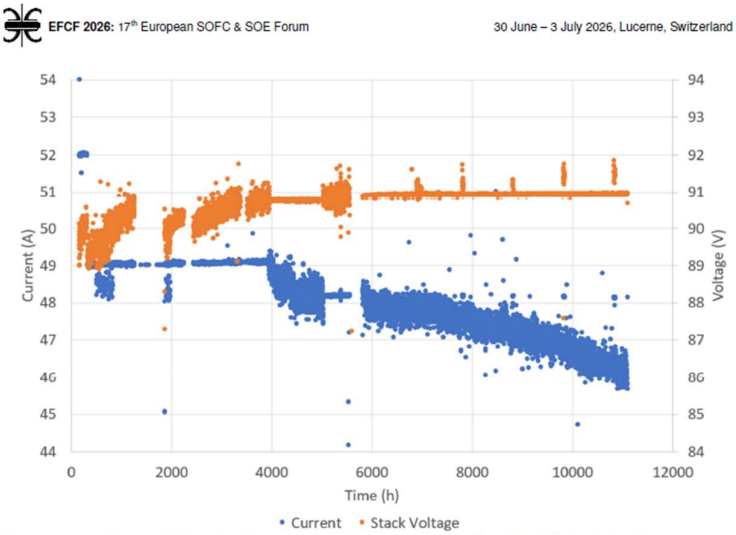

Figure 1. Evolution of the stack current and voltage during the 70-cell stack test.

In that work, the exact same stack design as the one used in our study was operated for 10,700 h under purely DC conditions at −0.6 A cm⁻², with 75–80% steam conversion at 700–720 °C. During the final 5,000 h, in potentiostatic mode, a degradation rate of −0.3 A k h⁻¹ was observed. This corresponds to approximately −0.6% k h⁻¹ relative to the initial current of 49 A (calculated on a current basis rather than on voltage). This degradation rate is approximately ten times higher than that observed in our work.

Although there are some small differences in operating conditions between the two studies, these recent DC results show that stable long-term operation is also achievable under DC conditions, but not at the exceptionally low degradation rates reported in our paper, particularly under galvanostatic operation, which places the stack under higher stress than potentiostatic mode. The comparison, therefore, suggests that the high durability observed in our study likely results from a combination of factors, including stack quality, system robustness, and test bench reliability, with AC:DC operation nevertheless playing a clear and important role in enhancing durability.

153-155 (173-178 in the updated draft): Unless I'm missing something, if standard thermoneutral operation, which effectively both stabilizes voltage (1.29 V/RU) and eliminates thermal gradients, does lead to monotonic degradation, then these cannot be the activators of dominant degradation pathways that AC/DC mode suppresses. Can the authors comment?

Authors: *We thank the reviewer for this insightful comment and agree that indeed, under potentiostatic operation, the cell voltage and temperature remain constant. In that mode, degradation is reflected in a gradual decrease in current density, leading to reduced H_2 production and an increased levelized cost of hydrogen (LCOH). However, AC:DC operation belongs to the broader class of (pseudo-)galvanostatic operating modes, where the voltage amplitude and frequency are adjusted to maintain a fixed average current density and, consequently, a constant H_2 production rate.*

Under conventional DC galvanostatic operation, degradation manifests as a progressive increase in the electrical energy/voltage input (under a given thermal energy supply) required to sustain the imposed current density. This increase in operating voltage can both further elevate the stack temperature through Joule heating and boost thermal degradation of the stack. Based on that, degradation under AC:DC operation would require a corresponding adjustment of the operating voltage amplitude and/or frequency to maintain the target current density. No such adjustment was required during the present test, particularly during the final ~22,000 h, when the stack was operated exclusively in AC:DC mode.
Therefore, the stable operating conditions, together with the negligible evolution of stack voltage and impedance spectra, and the post-mortem microstructural observations, provide strong evidence that AC:DC operation effectively suppresses degradation processes that, under conventional DC operation, could emerge after a few thousand hours. Nevertheless, in the absence of an equivalent long-term DC reference test under identical conditions, we acknowledge that the present results demonstrate a strong association between AC.DC operation and exceptional stack stability rather than definitive proof of causality, and we modified the text accordingly.

196 (224-232 in the updated draft): I would say that the electrochemical response was indeed stable during AC/DC operation, but based on the results shared, I'm not sure the causality is clearly shown.

Authors: *We thank the reviewer for this important observation. The experimental results clearly demonstrate highly stable electrochemical behavior over approximately 25,000 h of operation, with negligible changes in stack voltage, impedance spectra, and post-mortem microstructural features. Based on the arguments mentioned in the responses above, our observations are consistent with AC:DC operation phenomenologically contributing to the observed stability, but the absence of an equivalent long-term DC reference test under identical conditions challenges the definitive attribution of causality. However, as mentioned above, additional support regarding the beneficial effect of AC:DC operation is provided by recent results reported by Ouwelthes et al., "Steady-state operation of 70-cell SOEC stack – pushing the boundaries of long-term operation at low degradation rates" (EFCF 2026, A1201; no DOI currently available).*
We have therefore revised the manuscript to adopt more cautious language and to distinguish more clearly between the experimentally observed stabilization and the mechanistic interpretation of AC:DC operation. Nevertheless, besides the abovementioned, we would like to mention again that the beneficial effect of AC:DC operation becomes apparent in our experiment too, during the initial period and remains consistent with the exceptional long-term stability observed throughout the test.

222 (224-232 in the updated draft): The wide range of degradation rates found in the literature would suggest that materials, microstructures, manufacturing process,

operating parameters and quality of utilities (likely among others) all play a role in the stability (or lack thereof) of the stacks during standard DC operation. While it is very much warranted here, the comparison would be ideal if SolydEra had made no improvement to their cells over the past decade, and if utility quality was same during both tests. Both are likely not true.

Authors: *We thank the reviewer for this important comment. Indeed, the reported six-cell stack from SolydEra (ex SOLIDpower) operated under DC electrolysis cannot be regarded as an absolutely accurate comparable reference for the present study. As the reviewer correctly notes, SOEC degradation is influenced by numerous factors, including cell materials, microstructure, manufacturing processes, operating conditions, gas purity, thermal history, utility quality, and ongoing technological improvements. Consequently, differences between the two tests cannot be attributed solely to the operating mode. However, additional support for our reasoning in the previous comments regarding the beneficial effect of AC:DC operation is provided by recent results reported by Ouwelthes et al., "Steady-state operation of 70-cell SOEC stack – pushing the boundaries of long-term operation at low degradation rates" (EFCF 2026, A1201; DOI currently not available). A separate publication is under preparation.*
To address this concern, we have revised the manuscript to clarify that the previously reported DC stack is used as a contextual benchmark rather than as a direct control experiment. The comparison is intended to provide perspective on the magnitude of the stability observed in the present AC:DC-operated stack, while acknowledging that the quality of the stack and the test bench may also contribute to the observed differences.

287: Before jumping into economics, it would be quite interesting to discuss the limitations of the ACDC strategy. For example, the strategy decreases the maximum SC achievable, a parameter with high impact on efficiency (even assuming free/low cost vaporization). In your case, the 76% SOEC SC goes down to 63% overall. Describing the non-achievable window of SOEC operation would be a great addition, as well as the (module) efficiency cost. Similarly, how could the control parameter of the strategy evolve with degradation (even if low)? Voltage increase over time means that the stack will be less endothermic in SOEC, and becomes exothermic overall. Conversely, could this strategy help with heat generation in SOFC? And finally, what are the implications on large systems power electronics specifications? Is it limited, with current solutions being (somewhat) compatible? Should a new generation of solutions be developed?

Authors: *We thank the reviewer for this insightful comment and agree that the proposed strategy may narrow the achievable SOEC operating window, which must be balanced against the observed stabilization benefits. However, defining such a non-achievable operating window would require dedicated experiments across multiple operating conditions to separate the effects of frequency and amplitude on degradation, particularly at lower temperatures, where all processes become slower. Some relevant results in this direction can be found in* https://doi.org/10.1016/j.jpowsour.2026.239904 .

As degradation increases the cell voltage, SOEC operation becomes progressively less endothermic and may eventually become exothermic, thereby affecting the thermal balance and the effective control window over time. In the present work, this effect is accounted for through the lifetime definition adopted in the techno-economic analysis: end of life is reached when the calculated stack outlet temperature attains the thermal limit of 800 °C.

Accordingly, the balance-of-plant components, including heat exchangers, compressors, heaters, gas processing units, and power electronics, were sized based on end-of-life rather than beginning-of-life conditions. This ensures that the system can accommodate the increased heat generation and electrical demand associated with degradation. Furthermore, the degradation cost values already include both stack replacement and the additional electricity consumption resulting from efficiency losses over time. In particular, the power electronics are sized according to the maximum electrical demand at the end of the stack's life. While a purely DC-based operating strategy might also provide an acceptable lifetime, AC:DC offers an important additional benefit at the process level: it enables a constant hydrogen output and a fixed temperature profile throughout the plant lifetime. More generally, AC:DC operation appears to support a stable SOE stack throughout the entire lifetime. Rather than compensating for degradation by increasing temperature or lowering current density, the operator can maintain a fixed temperature—chosen according to component lifetime constraints—and apply an AC:DC current slightly below the thermoneutral point. This is particularly advantageous for integration with downstream processes, especially those that benefit from an initially endothermic operating point, and may also improve the expected round-trip efficiency of the system.

Whether AC:DC is necessary compared with DC operation cannot be answered in general terms, as it depends strongly on the specific plant design and whether the benefits of AC:DC justify the added complexity of the power electronics. Nevertheless, the present results improve the understanding of SOEC operation, strengthen the basis for techno-economic assessment, and help de-risk future investments in the technology.

The broader question of whether AC:DC operation may motivate the development of new converter concepts remains an important topic for future work and was not explicitly addressed here, as it would require a broader and more interdisciplinary investigation. Finally, while the potential regulation of SOFC heat generation is indeed an interesting implication, a detailed assessment would require dedicated thermal and control analyses and therefore lies beyond the scope of the present study.

299 (342-344 in the updated draft): Could the authors expand on their lifetime definition? Is it a maximum voltage / minimum acceptable conversion efficiency? If so, as long as the stack and its tightness are functional, what is stopping the operator to increase the temperature and expand the lifetime? Also, putting forward a 350+ kh projected lifetime likely calls for a sentence or 2 about aging of the tightness solution. The results here do not cover the stability of the glass ceramic matrix, and the vast majority of long-term tests end due to tightness failure.

Authors: *We thank the reviewer for this important comment and agree that the lifetime definition should be clarified more explicitly.*
In the present analysis, lifetime is not defined directly by reaching a maximum voltage or a minimum acceptable conversion efficiency. Instead, under galvanostatic operation, voltage degradation increases the electrical energy input required to maintain the imposed current density, thereby increasing heat generation and progressively shifting the thermal balance of the stack. Therefore, in this work, the end of life is defined as the point at which the calculated outlet temperature reaches 800 °C, which is taken as the maximum allowable operating temperature.
As the reviewer correctly points out, one possible operational strategy is to increase the stack temperature to compensate for the voltage increase and thereby maintain performance for longer. However, such an approach would also be constrained by the maximum allowable thermal conditions of both the stack and the overall system. In principle, the projected lifetime could be extended if a higher thermal limit were assumed. Nevertheless, this would require careful consideration of stack-level limitations, including sealing stability, glass-ceramic aging, mechanical robustness, and other degradation risks. It would also affect the design of the balance-of-plant, including heat exchangers and other thermal-management components, which would need to remain compatible with the higher operating temperature.
We also agree with the reviewer that projected lifetimes above 350 kh should be interpreted with caution, particularly regarding sealing durability. The present results do not explicitly assess the long-term stability of the glass-ceramic sealing matrix or the solution's tightness. These remain important practical limitations, as long-term failure is often associated with sealing degradation rather than only with electrochemical performance loss. We have therefore clarified in the manuscript that the reported lifetime is a model-based projection within the assumed thermal operating limits, and that additional validation would be required to confirm the long-term durability of the sealing system under such extended operation.

327-363, 347 in particular, and overall:
This works shows early degradation in ACDC mode, and no degradation throughout a later short DC operating window. I don't believe the latter result is properly discussed, given its massive implication. Regarding the statement of 347, this can only truly be known by switching a DC operated stack that degrades significantly to ACDC operation and showing some form of cancelation. Also, using a SolydEra stack for your demonstration does not really work in your favor, since their cells/stacks do show great stability even when DC operated. Also, 347 can only be stated if/when your results are compared to an equivalent DC test, which you do not have. Selected use of conditional might be warranted. The outstanding and impactful nature of the work done shouldn't be lessened in any way if/when DC data of SE stacks showing low degradation is published.

Authors: *We thank the reviewer for this thoughtful comment and agree with the points raised. As discussed in our responses to Comments 143 and 147, the present study does not report a matched long-term DC reference experiment on an equivalent stack under identical operating*

conditions. Consequently, we agree that the current specific dataset does not allow us to demonstrate causality or to conclude that AC:DC operation alone is responsible for the observed durability. However, across the SOEC literature, DC operation at around 750 °C has generally not, by itself, been sufficient to deliver the level of durability and performance observed here, with signs of Ni migration or YSZ electrolyte degradation being reported. In the present study, we report not only the longest SOEC stack test to date, but also the lowest degradation rate observed at stack level, with suppressed Ni migration and not observable degradation of the 8YSZ electrolyte. Notably, approximately 23,000 h of the total 25,000 h test duration, corresponding to about 90% of the experiment, were conducted under AC:DC operation. We agree that no measurable degradation was observed during the DC period in phase 5. However, this period lasted just 600 h and was followed by extended AC:DC intervals, which appeared to markedly reduce the voltage degradation rate, from 11.5% to 0.6%.

For this reason, we do not consider it equivalent to an independent long-term DC control experiment. We would also like to mention again that additional support for the beneficial impact of AC:DC is provided by recent unpublished work presented by Ouweltjes et al. at EFCF 2026 (abstract A1201), which reports steady-state operation of a 70-cell SOEC stack at low degradation rates. In that work, the exact same stack design as used in our study was operated for 10,700 h under purely DC conditions at −0.6 A cm⁻², achieving 75–80% steam conversion at 700–720 °C. During the final 5,000 h, in potentiostatic mode, a degradation rate of −0.3 A k h⁻¹ was observed. This corresponds to approximately −0.6% k h⁻¹ relative to the initial current of 49 A (calculated on a current basis rather than on voltage). This degradation rate is approximately ten times higher than that observed in our work.

However, in response to the reviewer's comment, we have revised the manuscript to use more cautious language throughout the Discussion and Conclusions. AC:DC operation is not presented as undoubtedly the demonstrated sole cause of the stack's durability. The comparison with previous SOEC durability studies is intended as contextual benchmarking and not as definite proof of causality.

The principal finding of the present work remains the experimental demonstration of record-stable operation of a 70-cell industrial SOEC stack, predominantly under AC:DC conditions, for 25,000 h, including more than 22,000 h with an average degradation rate of only 0.05 % kh⁻¹. Importantly, this result was achieved using a conventional SOEC fuel electrode architecture comprising Ni–YSZ fuel electrode, an 8YSZ electrolyte, a CGO barrier layer, and an LSCF–CGO oxygen electrode, without the introduction of novel cell materials.

Reviewer #2 (Remarks to the Author):

Dear authors,
thank you very much for this interesting and impressive paper about SOEC long-term operation.

I have only some minor comments/questions, please refer to the pdf attached.

Authors: 110: Addition of the word, ‘’ cell microstructural’’, as suggested by the reviewer; 146: Addition of the phrase, ‘’ cell microstructures’’, as suggested by the reviewer.

241-249

Statement solely on "optical" eye information or measured? From my eye-viewpoint a certain but not pronounced higher porosity could be seen after the operation. Optical fraction analysis would be great.

Authors: *We thank the reviewer for this valuable comment. The statement is not based solely on visual inspection, although we note that both the Ni percolation maps (green phase) and the porosity maps (black phase) presented in the main text qualitatively indicate already the presence of larger Ni particles and smaller porous clusters after testing, respectively. The fact that these features are observed relatively homogeneously throughout the analyzed microstructure, without a pronounced loss of Ni percolation or a significant increase in porosity near the electrolyte interface, provides important qualitative evidence of Ni coarsening while suggesting only a limited extent of Ni migration across the analyzed region. Furthermore, the Ni percolation maps obtained at the outlet side likewise do not show evidence of a pronounced loss of Ni percolation adjacent to the electrolyte interface, further supporting the conclusion that the observed microstructural evolution is relatively homogeneous across the electrode and not associated with substantial Ni migration away from the electrolyte. To quantitatively assess porosity evolution, we performed area-pore-fraction analysis of the AsB-SEM images using ImageJ, as described in Supplementary Note 7 and shown in Figs. S7–12. Multiple images acquired at different magnifications were analyzed before and after testing to minimize the field-of-view bias.*

Did you check whether the pore size fractions of FE changed after testing. It seems that pores become larger, but maybe overall not more.

Authors: *We thank the reviewer for this helpful comment. In the Supplementary Information (Supplementary Note 7, Figs. S7-S8), we have already included AsB-SEM-based pore-fraction analysis at multiple magnifications in the inlet region of Ni-YSZ fuel electrode (where the highest degree of degradation is expected), together with ImageJ segmentations used for the quantification.*
The analysis shows that, after 25,000 h of operation, the Ni–YSZ fuel electrode is slightly less porous than the untested reference, consistent with thermally activated, steam-promoted Ni coarsening. Locally, some regions adjacent to the electrolyte where the effect of Ni depletion is expected to be significantly more pronounced appear just slightly less porous, while a few larger pores randomly distributed along the interface can increase the measured area pore fraction. This indicates that the apparent increase in porosity is not primarily due to a homogeneous increase in the number of pores, but rather to limited Ni coarsening and Ni migration, as well as the presence of larger, isolated pores. Importantly, the differences remain small, supporting the conclusion that Ni migration and severe fuel-electrode degradation were substantially suppressed during the 25,000 h test.

Is figure d part from figure c? If yes could you indicate in figure c the area?

Authors: *We thank the reviewer for this comment. Figure d is indeed a higher-magnification image acquired from the same general region shown in Fig. c. However, the two images were obtained during different but consecutive SEM sessions, making it challenging to identify and mark the exact location of the higher-magnification image within Fig. c with confidence. Based on the acquisition approach used during the analysis, the region shown in Fig. d is expected to*

originate approximately from the central part of Fig. c. This location was intentionally selected to ensure that the image remained within the inlet region and was representative of the average microstructural features and degradation phenomena occurring in that area.

But if those "darker spots" are not composed of Cr, Al or Si, what are they? Pores or have you checked for Mn?

Authors: *We thank the reviewer for this valuable comment. The darker features observed in the micrographs are most likely associated with pores and/or minor surface imperfections introduced during sample preparation, such as small polishing scratches. To investigate their nature, EDS analyses were performed with particular attention to impurity species that could reasonably be expected based on the stack materials, operating conditions, and testing environment, namely Cr, Al, and Si. No enrichment of these elements was detected in the darker regions. EDS analysis did not reveal any detectable Mn peak, and significant Mn diffusion into the Ni phase would not be expected in the Ni-YSZ fuel electrode under the operating reducing conditions of this test. Furthermore, no substantial Mn source was present in the stack materials or the test setup that could plausibly account for Mn accumulation in the fuel electrode. We have clarified this point in the revised manuscript.*

Reviewer #3 (Remarks to the Author)

The manuscript describes a novel operating AC:DC strategy for minimizing degradation of SOEL stacks. The manuscript has a very high quality and is well written. The methodology and results are scientific, well discussed and sound. The established literature in this topic is up-to date and good referenced. The conclusions are underlined trough different evaluation methods (long-term operation, EIS and post-mortem analysis). The results are important and significant for the SOEL community.

Minor review recommendations are:

Format: use fixed blanks between numbers and units.

Authors: *We thank the reviewer for this helpful note. The issue is fixed.*

2) Format: The fond size of figures S14a and S17a is very small. Please increase.

Authors: *We thank the reviewer for this helpful note. The issue is fixed.*

3) The structure of the manuscript is separated into several sections (main text, methods, figure captions, and supplement), which are not logically ordered. Hence, it is very difficult to read the manuscript. It is recommended for the final manuscript to merge the information in logical sections.

Authors: *We thank the reviewer for this helpful comment. We believe that the manuscript follows a logical progression from general observations to increasingly detailed analyses.*

The first section presents the overall stack performance and the record-long-term durability in terms of voltage evolution. This is followed by impedance spectroscopy (EIS) analysis, which provides a deeper understanding of the electrochemical processes contributing to the observed voltage stability and enables the deconvolution of the degradation behavior into its individual components. Subsequently, SEM and EDS analyses are presented to investigate the microstructural and compositional evolution of the stack materials and, more specifically, to provide physical insight into the changes inferred from the EIS analysis and to better understand the origin of the observed impedance evolution. Finally, the techno-economic assessment is introduced to place the scientific findings into an industrial and commercial context.
Therefore, the manuscript is intentionally structured to progress from system-level performance to electrochemical analysis, to microstructural characterization aimed at interpreting electrochemical observations, and ultimately to industrial impact. We believe that this organization provides a coherent narrative that guides the reader from the principal experimental observations toward the underlying degradation mechanisms and their practical implications.

4) The degradation of the stack is mostly only given as relative value. Please also add the absolute degradation per cell in mv/kh.

Authors: *We thank the reviewer for this helpful suggestion. To facilitate comparison with previously reported SOEC degradation studies, we have now explicitly included the average absolute per-cell degradation rate. The stack voltage increased from 85.2 V to 86.2 V over approximately 22,000 h of operation, corresponding to a total increase of 1.0 V. For a 70-cell stack, this translates to an average voltage increase of approximately 14.3 mV per cell, yielding an average degradation rate of approximately 0.65 mV kh^{-1} per cell. This value has been added to the manuscript (line 25).*

5) Please describe the thermocouple positions and the calculation of the nominal average stack temperature more clearly.

Authors: *We thank the reviewer for this comment and agree that the thermocouple positions and temperature definition should be described more clearly. In the revised manuscript, we now specify that the thermocouples were positioned in-line in the gas paths, directly at the interfaces between the gas manifolds and the stack, such that temperatures were recorded at both the fuel-electrode (FE) and air-electrode (AE) inlets and outlets.*

We further clarify that the reported nominal temperature difference across the stack was calculated as:

$$\Delta T_{nom} = \frac{\left(T_{FE,out} + T_{AE,out}\right) - \left(T_{FE,in} + T_{AE,in}\right)}{2}$$

This quantity therefore represents the average gas temperature increase across the stack, based on the measured inlet and outlet temperatures on both the fuel-electrode and air-electrode sides.

6) The stack performance at the beginning of operation is not reported in the manuscript. Please give the important initial stack data: electrolysis power, steam conversion rate, electrical stack efficiency and produced flow rate of H_2.

Authors: *Thank you for this important comment. We agree that the initial stack performance parameters are important and have now included them in the revised manuscript.*
At the start of operation, the stack was operated at 40 A (-0.5 A cm^{-2}) and approximately 81.7 V, corresponding to an electrical power input of ~3.3 kW. This operating point resulted in a hydrogen production rate of approximately 2.5 kg day^{-1} (~0.104 kg h^{-1}), corresponding to about 3.5 kW on an LHV basis, assuming 100% Faradaic efficiency. Based on these values, the electrical stack efficiency was approximately 105–107% (LHV). The steam conversion was 63%, and the inlet gas composition included 10% H_2. These data have now been added to the main manuscript (Methods, Test bench and operating conditions: lines 441-446).

7) Usually, the degradation at the beginning of operation is higher compared to extended operation periods. Please discuss the impact of time on the low degradation in context to the impact of AC:DC method.

Authors: *We thank the reviewer for this insightful comment and agree that degradation rates in SOECs are often highest during the initial period of operation before decreasing as the stack undergoes conditioning and microstructural stabilization. Exposure to high temperature and steam content during the early stages of operation can introduce thermal and mechanical stresses throughout the stack. In Ni–YSZ FEs, these conditions, among others, promote Ni coarsening, leading to a reduction in triple-phase boundary density and a corresponding deterioration in electrochemical performance.*
From a thermodynamic point of view, phenomena such as Ni coarsening, Cr depletion/deposition, material softening, and thermal expansion are expected to gradually approach a stable equilibrium state during operation under steady temperature and gas compositions/flows. These degradation processes may therefore eventually reach a quasi-steady state if no additional driving forces are imposed. However, when an electrical bias is applied to the cell, particularly under galvanostatic conditions, the system is continuously driven away from equilibrium. In this case, the progressive increase in the voltage required to sustain the imposed current density can further increase local overpotentials and maintain the driving force for electrochemically induced degradation mechanisms, such as Ni migration and impurity deposition/poisoning (e.g., Si-containing species).
The combined action of thermally and electrochemically driven degradation processes can progressively increase cell overpotentials, which may ultimately promote impurity diffusion and the gradual detachment of Ni from YSZ, phenomena that have been phenomenologically correlated with accelerated increases in ohmic resistance. It has been shown that such

degradation can shift the electrochemical reaction zone from the inlet toward the outlet of the fuel electrode leading to further continuous extensive degradation of the Ni-YSZ fuel electrode and increase of voltage

Therefore, long operating times alone do not necessarily imply stable operation, particularly under galvanostatic conditions. Under potentiostatic operation, the cell voltage remains constant while degradation is reflected by a progressive decrease in current density and, consequently, a reduction in hydrogen production rate. This reduction in productivity would be expected to increase the levelized cost of hydrogen (LCOH) over time. As a result, stable voltage operation does not necessarily imply stable hydrogen production or stable and favorable economics. In this sense, potentiostatic operation is more likely to maintain a stable voltage, but at the expense of reduced hydrogen output and less favorable economic performance.

In the present study, the exceptionally low voltage drift, stable impedance response, and limited microstructural evolution observed over approximately 22,000 h of operation under the pseudo-galvanostatic AC:DC mode suggest that the above-mentioned electrochemically driven degradation pathways remained strongly suppressed. While the absence of an equivalent long-term DC reference test prevents a definitive separation of time-dependent stabilization effects from those associated with AC:DC operation, the results are consistent with AC:DC operation mitigating degradation mechanisms such as Ni migration and impurity accumulation, thereby allowing the electrode microstructure to evolve toward a more stable long-term state.

8) Please shortly specify/discuss the 5 different electrochemical processes identified in the DRT spectra.

Authors: *We thank the reviewer for this valuable comment. We agree that a brief discussion of the five DRT processes improves the interpretation of the electrochemical results, and we have added a corresponding description to the revised manuscript.*

For fuel-electrode-supported SOECs comprising a Ni–YSZ fuel electrode, 8YSZ electrolyte, CGO barrier layer, and LSCF–CGO oxygen electrode, five DRT peaks are commonly reported in the literature. As a general rule, the highest-frequency processes are associated with the fastest electrochemical phenomena, whereas the lower-frequency processes are typically related to slower gas transport, gas conversion, and, more generally, mass-transfer phenomena. Nevertheless, the exact summit frequency of a given process depends on the electrode microstructure and operating conditions. Therefore, definitive peak assignment would require a dedicated initial EIS characterization campaign under systematically varied operating conditions.

Based on literature reports, the sensitivity of the processes to gas composition (Fig. S4), and post-mortem microstructural analysis, the evolving high-frequency process, P2 (associated with the R2 resistance), is most likely related to charge-transfer processes in the Ni–YSZ fuel electrode. This interpretation is consistent with the observed Ni coarsening, which is expected to occur under prolonged exposure to high temperature and high steam content. Such coarsening reduces the triple-phase boundary density and is therefore expected to increase the corresponding polarization resistance.

The P4 process (associated with the R4 resistance) contributes significantly to overall polarization losses but exhibits a considerably more stable evolution than P2. Owing to its lower-frequency nature and its sensitivity to the H_2/H_2O ratio, we tentatively assign this process to fuel-electrode gas-conversion phenomena. The lowest-frequency process, P5 (associated with the R5 resistance), is likely related to gas diffusion and mass-transport limitations. The relatively stable medium-frequency process, P3 (associated with the R3 resistance), is expected to be related to the stable performance LSCF–CGO oxygen electrode, whereas the high-frequency process, P1 (associated with the R1 resistance), may be related to oxygen-ion transport within the Ni–YSZ electrode structure.
However, the evolution of R1 and R3 exhibits considerable scatter, making definitive conclusions regarding the exact nature of these two processes challenging.

9) According to Fig. 2d the contribution of R4 is in the same range as Rs and even higher compared to R2. Please comment the contribution of R4.

Authors: *We thank the reviewer for this important comment. We agree that R4 represents a substantial contribution to the overall stack polarization. Based on its low-frequency nature, its sensitivity to the H_2/H_2O ratio, and the literature on Ni–YSZ fuel-electrode-supported SOECs, R4 is most likely associated with gas-conversion-related processes in the Ni–YSZ fuel electrode.*
The relatively large magnitude of R4 is expected for this SOC architecture, as the active fuel electrode is relatively thick (~260 µm). Under these conditions, CSTR concentration gradients (https://doi.org/10.1149/1.1838654) within the fuel electrode can contribute significantly to the gas-conversion resistance, in addition to the PFR concentration gradients along the flow direction (https://doi.org/10.1149/1.3116247, https://doi.org/10.1149/1.3050402 , https://doi.org/10.1149/ma2008-01/10/431). Thus, both through-thickness concentration gradients and plug-flow-type gas conversion along the channel are expected to contribute to the measured R4 response.
Importantly, R4 remains comparatively stable over the ~22,000 h AC operating period. We interpret this stability as an indication that the Ni–YSZ fuel-electrode microstructure, including the inlet region, did not undergo severe degradation during operation. Significant degradation of the inlet region would be expected to alter local reaction distributions and gas-conversion gradients, thereby affecting the evolution of R4.

10) The cost of stack is given in €/m^2, which is quite unusual. Should it not be €/kW or €/MW? Please comment. (see also comment in Supplement Table S8).

Authors: *We thank the reviewer for this comment. We agree that stack costs are frequently reported in €/kW or €/MW in the SOEC literature. In the present work, however, we expressed the stack cost in € m^{-2} of active area, because this basis is more directly related to the physical amount of stack hardware required and is therefore more transferable across different operating conditions.*
The main reason is that a €/kW-based cost depends on the selected operating point. For a given stack technology, the electrical power delivered per unit active area varies with current density

and cell voltage. As a result, the same physical stack can correspond to different €/kW values depending on how it is operated. In contrast, the cost of manufacturing the stack is more fundamentally associated with the amount of active cell area and the corresponding hardware, such as interconnects, seals, and assembly. For a plant designed for a fixed hydrogen production rate, hydrogen production is proportional to the total stack current, i.e., the product of current density and active area. Therefore, if the current density is reduced from 1.0 to 0.5 A cm^{-2}, the active stack area must approximately double to maintain the same hydrogen production rate. Since the physical amount of stack hardware also scales with active area, the stack cost should likewise be approximately double.
By contrast, the total electrical power does not scale in the same way. For a fixed hydrogen production rate, the total current remains approximately constant, while the electrical power depends on the product of total current and operating voltage. Although lower current density may reduce the cell voltage because of lower overpotentials, the voltage does not decrease proportionally with current density. Therefore, expressing stack cost directly in €/kW can be misleading when translating costs between operating conditions, because it does not necessarily capture the corresponding change in required stack area.
Using an area-based cost formulation, therefore, allows the cost estimate to remain linked to the physical stack size needed to achieve a given hydrogen production target. It also avoids introducing inconsistencies when comparing SOEC and SOFC operation, where differences in power density and voltage would otherwise make a €/kW-based value strongly dependent on the selected operating point.
To avoid ambiguity, we have expanded the explanation of this assumption in the revised manuscript and in Supplementary Table S8.

11) Add doi numbers of references (if available).

Authors: *We thank the reviewer for this helpful note. The issue is fixed.*

Reviewer #4 (Remarks to the Author)

This is an interesting work, which advances the SOFC development. I suggest the publication of this work after considering the following comments.

1. There is a transient behavior before the stabilization (the first 3000 hours). What happens during this period? A more detailed analysis during this period may help the readers' understanding.

Authors: *We thank the reviewer for this helpful comment. We have added a more detailed discussion of the first ~3000 h of operation in the main draft (lines 147-156), Supplementary Information, Note 3, Table S2 and Fig. S2.*
During the first ~2,700 h of operation, multiple operating conditions, including DC and AC:DC modes, different temperatures, and current densities, were systematically investigated to assess their influence on stack performance and identify suitable long-term operating conditions. This period reflects not only electrochemical conditioning, but also equilibration of system-level components and boundary conditions, including preheaters, metallic components, and parameters such as air humidity, temperature, and pressure. Following this optimization phase,

the stack was operated under AC:DC mode and entered a stabilized operating regime, in which the average voltage degradation rate remained as low as 0.05 % kh^{-1} over the remaining ~22,000 h of the 25,000 h test. During this initial period, The first DC period at 730 °C and 0.50 A cm^{-2} exhibited the highest apparent degradation rate, with the stack voltage increasing from 81.4 to 82.7 V over 138 h, corresponding to a degradation rate of 9.4 V kh^{-1}. Following the transition to AC operation at 750 °C and the same current density, the degradation rate decreased to 0.7 V kh^{-1}, corresponding to a reduction of approximately 93%. Although the degradation rate increased during the subsequent operation at 700 °C (5.8 and 4.6 V kh^{-1} for 0.50 and 0.20 A cm^{-2}, respectively), it remained below the value observed during the initial DC period. Upon returning to AC:DC operation at 750 °C, the degradation rate further decreased to 0.5 V kh^{-1}, corresponding to approximately a 95% reduction relative to the initial DC period. These observations suggest that the beneficial effect of AC:DC operation became apparent already during the early stages of the durability test, well before the onset of the long-term stabilized regime. Nevertheless, the first ~3000 h should not be interpreted as a single steady-state degradation regime, as the stack experienced multiple changes in operating mode, temperature, and current density. In addition, this period likely includes conditioning effects arising from the initial exposure of the stack materials to high temperatures, steam, and electrochemical polarization.

2. The AC:DC waveform uses 30 Hz. Why this frequency is used? Some discussion and justification is needed.

Authors: *We thank the reviewer for this valuable comment. In the revised manuscript, we have clarified that the choice of 30 Hz was based on earlier AC:DC SOEC work and selected as a practical operating frequency (https://doi.org/10.1016/j.jpowsour.2022.231040).*
We further note that the later study by Mattera et al., published in 2026 after the present test had already started, provides supporting evidence for this choice (https://doi.org/10.1016/j.jpowsour.2026.239904). That study examined AC:DC frequencies between 0.1 and 1000 Hz and found that frequencies above 0.1 Hz yielded similar electrochemical performance, whereas 0.1 Hz resulted in measurable oscillations in outlet gas composition. Although this study was not the original basis for selecting 30 Hz in the present work, it supports the view that a frequency in this range is a reasonable trade-off between stable operation and practical implementation.
Because the purpose of the present study was to evaluate long-term durability rather than optimize AC:DC frequency, only a single frequency was used throughout the experiment. We have clarified this in the revised manuscript (lines 456-463), and Supplementary Note 2.

3. An averaged DRT analysis is given across all eight cell groups, which is interesting but also risks masking heterogeneity. I suggest the authors also show the DRT of each group.

Authors: *We thank the reviewer for this valuable suggestion. We agree that averaging can, in principle, mask heterogeneity between different cell groups. However, we note that a complete*

DRT analysis, including all DRT moments, is already provided in Fig. S4b for a representative cell group. In addition, the evolution of the voltage and individual Nyquist plots for all eight cell groups is presented in Fig.S3 and Fig. S5, respectively, allowing the degree of heterogeneity across the stack to be assessed.

As shown in both Fig. S3 and Fig. S5, all cell groups exhibit very similar voltage and impedance patterns and temporal evolution, indicating a high degree of electrochemical uniformity across the stack. The main exception is Group G5, which exhibits a distinct high-frequency response in Fig. S5. Based on its frequency characteristics, its persistence throughout the test, and the existing SOEC literature, we consider this feature most likely related to the electrical wiring and cabling associated with that specific group rather than to a difference in cell degradation behavior.

Given the large number of spectra and DRT-derived plots that would be generated, we believe their inclusion would substantially increase the volume of supplementary material while providing limited additional insight beyond that already available from the individual Nyquist plots and the representative DRT analysis. For this reason, we have retained the current presentation, which we believe provides a balanced assessment of both the average stack behavior and the degree of heterogeneity among the cell groups.

4. The post-mortem analysis is mostly qualitative. Quantitative analysis, such as the particle size of nickel, porosity…., is suggested to be provided.

Authors: *We thank the reviewer for this valuable suggestion. While we agree that quantitative post-mortem analysis can provide important insight into degradation mechanisms, we note that quantitative porosity analysis has already been performed and is included in the Supporting Information (Supplementary Notes 6-7 and Figs. S6–S17).*

Specifically, area-pore-fraction analysis was conducted on AsB-SEM images in ImageJ to quantify porosity evolution before and after testing. Several images acquired at different magnifications were analyzed to minimize the field-of-view bias. The results indicate a measurable decrease in porosity following long-term operation, consistent with the qualitative observations from the porosity maps presented in the main text.

Regarding Ni particle-size analysis, we note that the primary objective of the post-mortem investigation was to evaluate whether the impedance evolution observed during operation could be associated with significant Ni migration. In this regard, both the Ni percolation maps (green phase) and the porosity maps (black phase) indicate that the microstructural evolution is relatively homogeneous throughout the analyzed Ni-YSZ fuel electrode region, without a pronounced loss of Ni percolation or major spatial variations in porosity near the electrolyte interface. Furthermore, the Ni percolation maps obtained at the outlet side likewise do not show evidence of a significant loss of Ni connectivity adjacent to the electrolyte interface.

Consequently, we do not expect a quantitative Ni particle-size distribution analysis to provide substantial additional information regarding Ni migration. Such an analysis would primarily quantify the extent of Ni coarsening, an expected degradation phenomenon that is already qualitatively evident from the Ni percolation maps, as evidenced by the presence of larger Ni

particles after testing. In contrast, significant Ni migration would be expected to manifest as a measurable depletion of the Ni network and a loss of percolation near the electrolyte interface, neither of which is observed in the present study.
For these reasons, we believe that the combination of quantitative porosity analysis and the spatially resolved Ni percolation maps provide sufficient evidence to support the conclusions drawn from the EIS analysis.

5. Although a short-stack DC test is compared, no long-duration DC control test on the same stack design is compared, making the direct comparison of the AC:DC control somehow weak. I can understand additional test on this issue will take too much time. If such a test is not feasible to do, some explanation is preferred.

Authors: *We thank the reviewer for this important comment and agree that the absence of a matched long-term DC control experiment limits the ability to directly isolate the effect of AC:DC operation.*
We note that the manuscript includes a comparison with a previously reported 10,700 h DC electrolysis test conducted on a stack from the same manufacturer. While this provides useful context and suggests that the durability observed in the present work is exceptional, we agree that the two studies cannot be considered directly equivalent. Differences in stack generation, operating conditions, test-bench infrastructure, and utility quality may influence the observed performance. Therefore, this comparison is intended only as a solid but conceptual benchmark rather than as a controlled reference experiment.
A dedicated long-duration DC control test on an equivalent stack would undoubtedly provide a more rigorous assessment of the specific contribution of AC:DC operation. However, given the objective of demonstrating stack durability under AC:DC operation over several tens of thousands of hours, and the risk that prolonged DC operation could accelerate degradation and compromise the long-term experiment, such a test was not considered feasible within the scope of the present study.
Also, given the current understanding of SOEC degradation under continuous DC polarization, the beneficial effect already observed during the early AC:DC periods of the present experiment, and the fact that the aim of this study was specifically to investigate stack durability under AC:DC operation over several tens of thousands of hours, a substantially longer DC operating period was considered unnecessarily risky. Continuous unidirectional polarization imposes sustained electrochemical, thermal, and mechanical gradients across the stack, increasing the likelihood that a local failure in a single cell could trigger accelerated degradation that propagates through the stack and compromises the long-term experiment.

Nevertheless, additional support for our reasoning in the previous comment regarding the beneficial effect of AC:DC operation is provided by recent results reported by Ouwelthes et al., "Steady-state operation of 70-cell SOEC stack – pushing the boundaries of long-term operation at low degradation rates" (EFCF 2026, A1201; no DOI currently available). For completeness, we attach the main figure from that work here but not in the main draft.

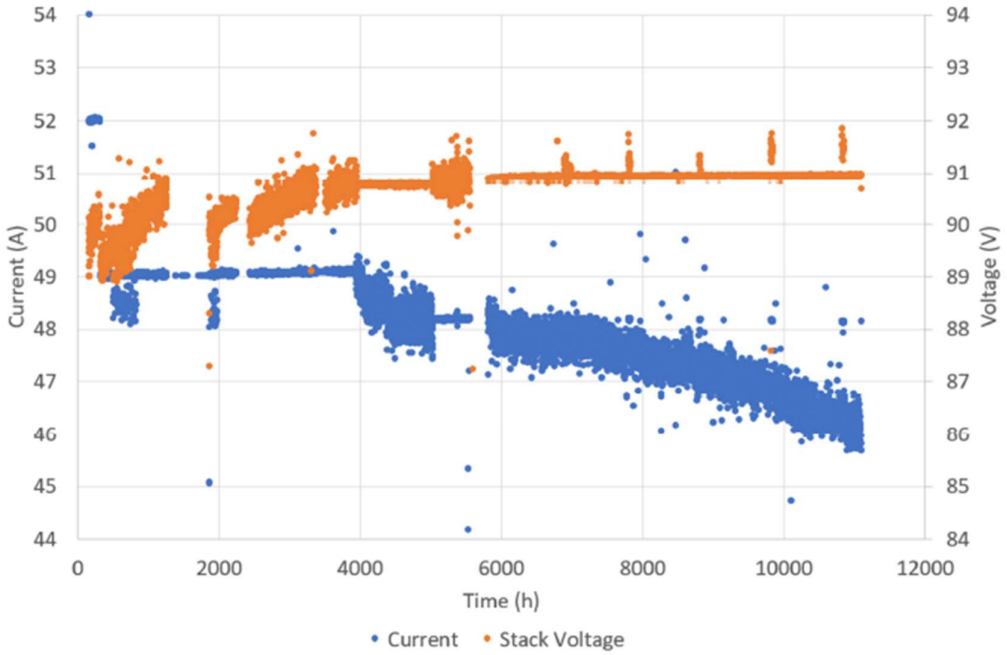

Figure 1. Evolution of the stack current and voltage during the 70-cell stack test.

In that work, the exact same stack design as the one used in our study was operated for 10,700 h under purely DC conditions at −0.6 A cm⁻², with 75–80% steam conversion at 700–720 °C. During the final 5,000 h, in potentiostatic mode, a degradation rate of −0.3 A k h⁻¹ was observed. This corresponds to approximately −0.6% k h⁻¹ relative to the initial current of 49 A (calculated on a current basis rather than on voltage). This degradation rate is approximately ten times higher than that observed in our work.

Although there are some small differences in operating conditions between the two studies, these recent DC results show that stable long-term operation is also achievable under DC conditions, but not at the exceptionally low degradation rates reported in our paper, particularly under galvanostatic operation, which places the stack under higher stress than potentiostatic mode. The comparison, therefore, suggests that the high durability observed in our study likely results from a combination of factors, including stack quality, system robustness, and test bench reliability, with AC:DC operation nevertheless playing a clear and important role in enhancing durability.

However, we have revised the manuscript to avoid attributing the observed durability exclusively and undoubtably to AC:DC operation and instead present the results as being consistent with a beneficial role of AC:DC operation in combination with other factors that might influence long-term stack durability.