General Response (GR)

GR-1: Thank you to the reviewers and the journal editor. We understand the limitations and accept transfer to ChemElectroChem (see response 1-2).

GR-2: In switching to an electrochemistry journal we propose a minor modification to the title, changing “catalysis” to “bioelectrocatalysis” arriving at the following: “A generalized kinetic framework applied to whole-cell bioelectrocatalysis in biofilm flow reactors clarifies performance enhancements”

GR-3: We have answered all questions posed by the reviewers. We resubmit the paper and SI in both clean and marked up format. The latter contains changes marked in green highlights.

Reviewer 1

Comment 1-1: A kinetic study in flow reactor has been reported employing Geobacter sulfurreducens in whole cells form. In my opinion the article is too specialised and lacks of the application and synthetic value that I would like to see for a journal such as ChemCatChem. I see more appropriate the submission to its sister journals ChemBioChem or ChemElectrochem.

Response 1-2: We understand that the approach is somewhat fundamental in that it reports bioelectrocatalysis without yielding a chemical product, as is typical for submissions to ChemCatChem. Therefore, we agree to transfer to ChemElectrochem where the reviewer (and editor) believes the material will be more relevant.

Comment 1-2: Anyway, the article is well written and structured and I have just minor comments for improvement:

Response 1-2: Thank you. See our point-by-point response below.

Comment 1-2.a: Update reference 11 (submitted)

Response 1-2.a: We have updated the reference (in its new position) to be:


Also, as this work is now accepted, we added the following sentence near the end of the communication explaining the opportunity to use this kinetic approach to study Geobacter biofilms which are in fundamentally different metabolic states.

“Characterization of these kinetic parameters may be useful to quantify the effect of external factors on EAB activity, such as flow-induced changes to metabolic state recently reported for Geobacter sulfurreducens biofilms at ultra-low Ac concentrations.**

Comment 1-2.b: Include microfluidic in the table of contents of supporting information (point 2)
Response 1-2.b: Done. Point 2 in the SI table of contents now reads: “Microfluidic device fabrication, anaerobic environment and inoculation”

Comment 1-2.c: Supporting information: clarify that aqueous solutions are used for the experiments, which in many cases is omitted (pages S4 and S10) or add information about the buffer (page S5 concentration and pH)

Response 1-2.c: We added this clarification to all relevant sections in the SI.

Reviewer 2:
Comment 2-1: This is a very good piece work. The manuscript is well written, which is recommended publishing after a minor alternation.

Response 2-1: Thank you. See our point-by-point response below.

Comment 2-2: whole-cell catalysis is very interesting. But please emphasize more on its definition and advantages.

Response 2-2: We added a new paragraph in the introduction (in green following this response). The advantages of whole-cell catalysis has been explained more in this section. Also, additional references were added to put readers in contact with the highly relevant literature in field of whole-cell catalysis.

“Bacterial biofilms are promising for the same reasons, including certain benefits making them candidates for industrial processes as well.6,7 These include their preference for surface attachment, making them ideal for heterogeneous catalysis; and their protective self-produced extracellular polymeric matrix, which can mitigate challenges related to toxicity, leading to long-term productivity even under harsh conditions.8,9 Promising areas of application include biodegradation and biochemical synthesis.10”

Comment 2-3: According to Fig 1, the working electrode is still much smaller than the flow cell. Ho do the authors justify "whole-cell"?

Response 2-3:
The reviewer is right, only a fraction of the microchannel is in contact with the electrode. We believe there is confusion about the use of “whole-cell” which does not refer to the flow cell. Confusion here is related to our use of the term “cell” for both the (microfluidic electrochemical) “flow cell” and (biological catalytic) “whole-cells”. To avoid this problem, we have removed all references to “flow cell” and, where possible, replaced them with “biofilm flow reactor”, which is also listed among the keywords (in the main paper and SI). We thank the reviewer, addressing this comment significantly improves the readability of the paper, in our opinion.

Comment 2-4: Fig. 2: how did the authors get the pulse-like i-t curves? Please specify the flow profile vs. time.
Response 2-4: The original Figure 2 was created from flow pulses using a computer control syringe pump. These changes were applied from 0.2 mL·h⁻¹ to an elevated flow rate (0.4, 0.6, 0.8, 1, 2 or 3 mL·h⁻¹) and then back down to the baseline flow rate before moving to the next elevated flow rate. To better explain the applied flow conditions that caused the measured biocurrent outputs, we have reworked Figure 2 to add Fig. 2a (below) which corresponds to the original Figure (now Fig. 2b). Minor changes to the main text and figure caption were made in relation to adding Fig 2a. See green highlights in the main paper.

Comment 2-5: Fig 3 b: I am not sure if 5 points were conducted for the green and red lines.

Response 2-5: In Figure 3b, each line is comprised of 5 points, arising from five different nutrient concentration of acetate. As higher acetate concentrations resulted in lower x-axis values, the concentrations 10 and 7 mM are at the lowest values along the x-axis. As these two concentrations result in similar current (I) values, (and therefore similar conversion, P) the data points and error bars overlapped at high concentrations. This effect was pronounced at high flow rates (red and green curves), but are indeed separate points. To clarify this point for the reader, we added a line in the Figure 3b caption:

"Each line is comprised of 5 points from the 5 concentrations in (a) which progressively became overlapping at low abscissa and ordinate values with increased Q."

Comment 2-6: More references are desirable.

Response 2-6: Five new references have been added to better guide readers in the areas of whole-cell catalysis and electron transfer in electroactive biofilms.

Also, the SI contains 4 additional references on biocatalysis in flow and electrochemical analysis of EABs.