Exploring the influence of intermittent irradiation on light-driven catalytic water oxidation – DoE based parameter optimization using automated data set generation and bayesian optimization

D. Ziegenbalg [1], D. Kowalczyk [1]

[1]Institute of Chemical Engineering, Albert-Einstein-Allee 11, 89081 Ulm/Germany

E-Mail presenting author: daniel.kowalczyk@uni-ulm.de

The development of new photocatalytic systems requires the screening of large parameter spaces to identify suited reaction conditions. This can be time consuming and coupled to an extensive experimental effort. Aiming at the utilization of sunlight to drive reactions, the intermittency of solar light intensity depicts a particular challenge. Therefore, operation under unsteady irradiation is relevant for technical application. Taking the photosensitized water oxidation catalysis (WOC) as an example, pulsed irradiation was realized hydrodynamically. Resulting in higher turnover numbers and frequencies under pulsed operation.[1] This could be traced to the deactivation of the used photosensitizer (PS) due to local accumulation of reactive species. To counteract deactivation, reaction and pulse operation conditions were varied systematically. Therefore, the mean photon flux, pulse length and duty cycle, were studied using the discussed WOC system. For comparability reasons the experiments were performed both under pulsed (PI) and unpulsed (UPI) irradiation. The relative final turnover numbers ($\mathrm{TON}_{\mathrm{final,rel}}$) and photonic efficiencies ($\xi_{\mathrm{final,rel}}$) were calculated as the fraction of the absolute TON of the PI and UPI setup. The results confirm a strong influence of pulsed operation, both in terms of $\mathrm{TON}_{\mathrm{final,rel}}$ and $\xi_{\mathrm{final,rel}}$. For specific operation conditions ($\bar{q}_{pulsed}$ = 0.5 µmol/s, t_{pulse} = 25 ms, DC = 25 %) the $\mathrm{TON}_{\mathrm{final,rel}}$ reaches the same value for pulsed as for unpulsed operation, using just 25 % of the photons. Therefore, intermittent irradiation must not only be considered with respect to solar light utilization but depicts a powerful reaction engineering tool to leverage the reaction performance. To screen the the extended parameter space, targeted pulse condition variation will be coupled to automated data set generation and bayesian optimization to identify optimal operation conditions.[2]

[1] M. Sender, F. L. Huber, M. C. G. Moersch, D. Kowalczyk, J. Hniopek, S. Klingler, M. Schmitt, S. Kaufhold, K. Siewerth, J. Popp, B. Mizaikoff, D. Ziegenbalg, S. Rau, ChemSusChem 2022, 15, e202200708.
[2] Y. Chen, K. Bi, C.-H. J. Wu, D. Ben-Arieh, A. Sinha, 2021, doi.org/10.48550/arXiv.2108.02289.