processes

MDPI

*Supplementary information*

# Improving the Treatment Efficiency and Lowering the Operating Costs of Electrochemical Advanced Oxidation Processes

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## Details about the GDE manufacturing process

The manufacture of the GDE through the "calandering" dry manufacturing process is depicted in Figure S1. First, the materials used, namely the carbon-based catalyst (Printex L6, Orion Engineered Carbons, Luxembourg) and PTFE (Dyneon, Germany), were mixed (IKA, Germany). The mixture was spreaded using powder spreading and doctor blades onto an Au-plated Ni mesh (mesh size: 0.5 mm × 0.5 mm; wire gauge: 0.14 mm) and calandered between two rollers to produce a compacted, firmly adhering active layer. Finally, the GDEs were sintered at 340 °C.

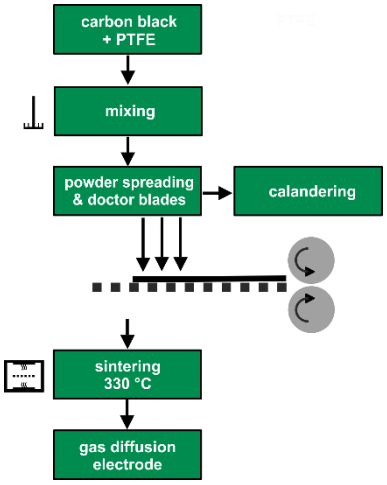



**Figure S1.** Schematic sequence of GDE production by calandering. The calandered GDEs were coated on one side of the substrate mesh.

## Details about the cell designs

The complete assembly of cell a is shown in Figure S2. The main components from the left to the right side are the stainless steel plate acting as the cathode withintegrated water distribution channels (diameter of 1.5 mm and distance of 4.3 mm), the seal for the defined electrode distance, the BDD anode, the backside seal of the anode, and the stainless steel end plate.

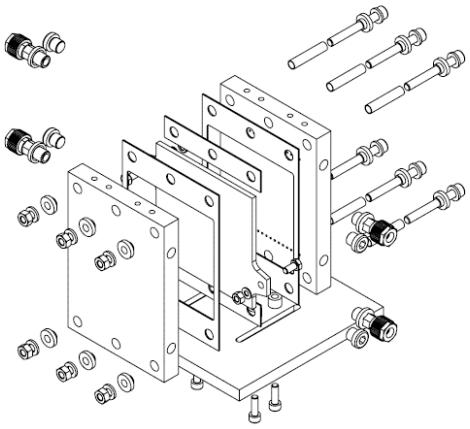
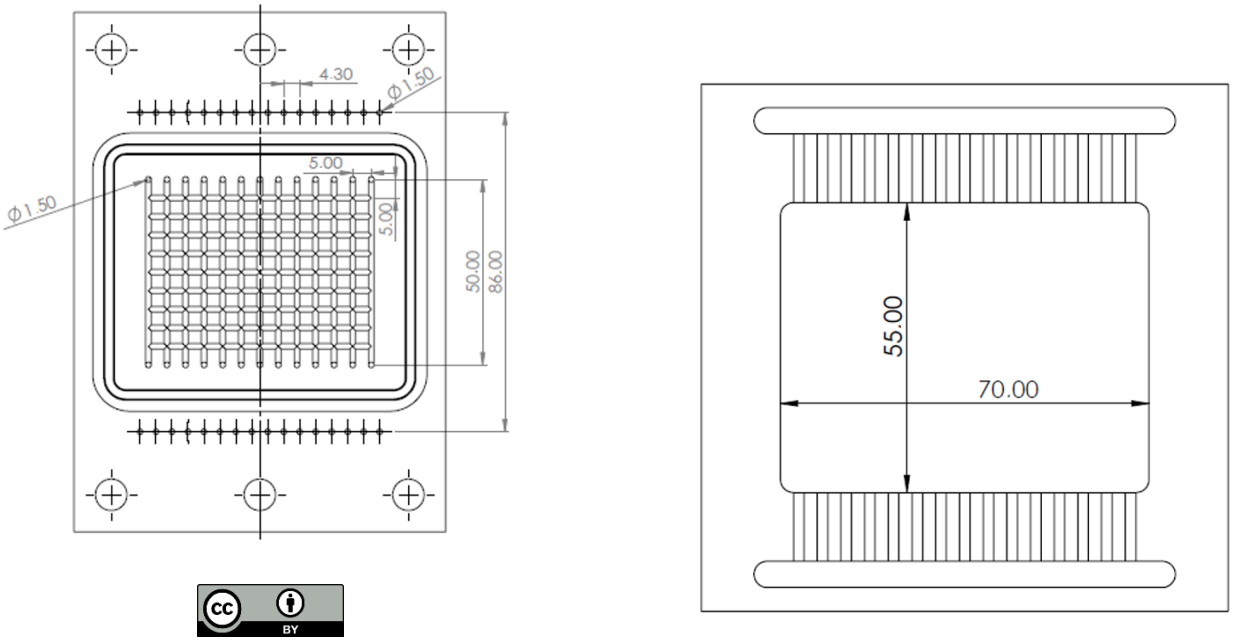


**Figure S2.** Exploded view of cell design a with a stainless steel cathode and a boron-doped diamond anode.

For cell design b, the most functional parts are the structured steel plate (Figure S3a), which has a dual function as an electrolyte distributor and feed system for gas and electrical connection of the GDE.

The structured flow field supplying the GDE has a width of 50 mm and a height of 12 mm. The diameter of the inlet and outlet holes for gas supply is 5 mm. The drilled structure has channels with a depth of 0.75 mm. The channels for water and gas supply for the piping have a diameter of 8.5 mm. In total, the steel plate is 15 mm thick. The PTFE frame (Figure S3 b) is 2 mm thick and the channels for water distribution are 2 mm wide with a depth of 1 mm, supporting each hole of the water inlet and outlet.






(**a**) (**b**)

**Figure S3.** The structured steel plate (**a**) and the PTFE frame (**b**) in cell design b.

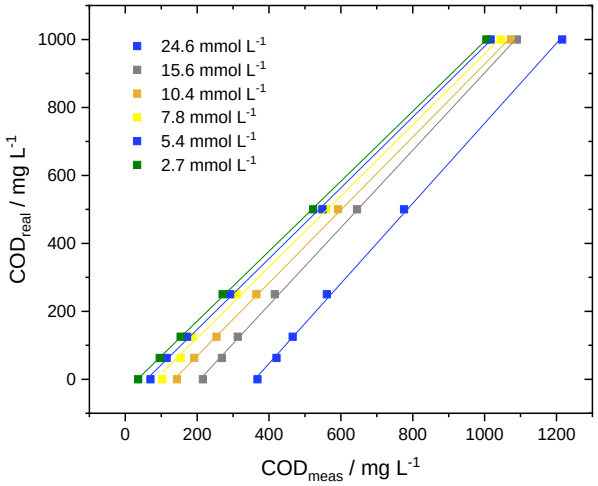



**Figure S4.** Correlation between the measured COD values, including an overdetermination due to $H_2O_2$ interference, and the real COD values for phenol in 9 g $L^{-1}$ $Na_2SO_4$ solution. Different $H_2O_2$ concentrations were investigated, which are given in the figure legend above.

**Table S1.** Equations for the linear fits from Figure S4. The coefficient of determination ($R^2$) was 0.9985 for the given equations.

| $H_2O_2$ Concentration / mmol $L^{-1}$ | Linear Fits |
|---|---|
| 24.6 | y = 1.1781 x − 424.79 |
| 15.6 | y = 1.1385 x − 236.64 |
| 10.4 | y = 1.0732 x − 145.72 |
| 7.8 | y = 1.0468 x − 88.314 |
| 5.4 | y = 1.0414 x − 61.544 |
| 2.7 | y = 1.0293 x − 34.765 |

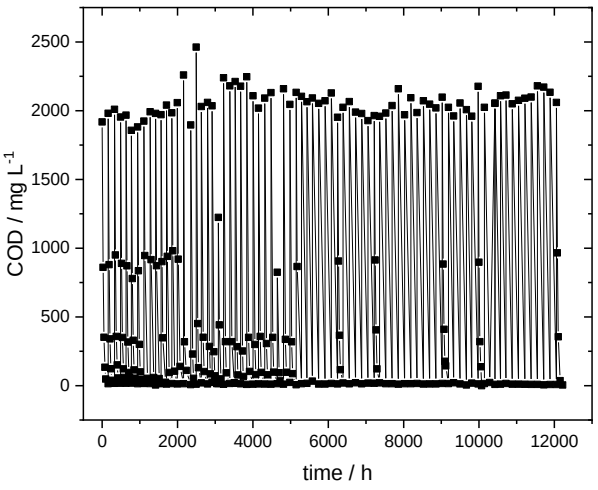



**Figure S5.** COD degradation in dependence of time during 77 batch treatments, with a total runtime of 12,222 h for cell type a.

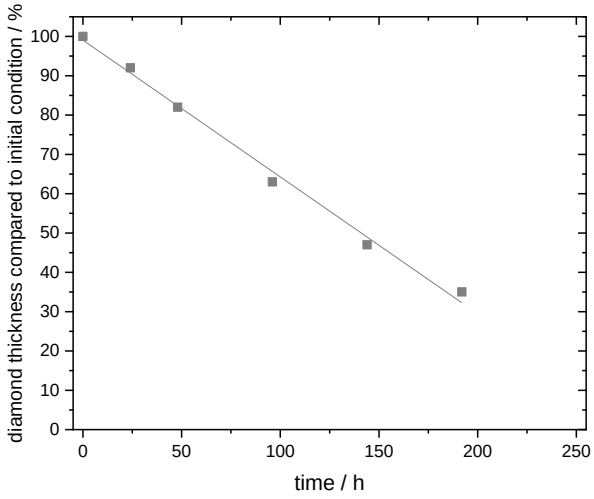



**Figure S6.** Durability test of a BDD anode. The diamond loss ranged between 6 and 10 µm. The coefficient of determination ($R^2$) of the linear regression was 0.994. The conditions for the accelerated corrosion test were in accordance with those published in [31,32] for diamond etching due to high current densities ($j$ >> 1 kA m$^{-2}$) and the degradation of an electrolyte of short-chain $CH_3$-containing molecules.

**Estimating the lifetime of the electrode**

To estimate the lifetime of the electrode, the diamond loss (measured by beta backscattering) was divided by the operational time for the DIACHEM® electrode in cell type a, according Equation (S1).

$$A\ [\mu m\ h^{-1}] = \frac{Loss\ of\ coating\ thickness\ [\mu m]}{Operation\ hour\ [h]} \quad (S1)$$

$$LT\ [h] = \frac{Coating\ thickness\ [\mu m]}{A\ [\mu m\ h^{-1}]} \quad (S2)$$

Based on the above, the diamond loss per hour A was calculated. Due to the data described in Figure S6, a linear diamond coating loss of 9 µm was set as the range for the lifetime estimation. The remaining 6 µm from a total coating thickness of 15 µm was not considered in the case study for reasons of data uncertainty. The lifetime LT is the quotient of the considered coating thickness of 9 µm and factor A (Equation (S2)).