Combined Anodic and Cathodic Peroxide Production in an Undivided Carbonate/Bicarbonate Electrolyte with 144 % Combined Current Efficiency

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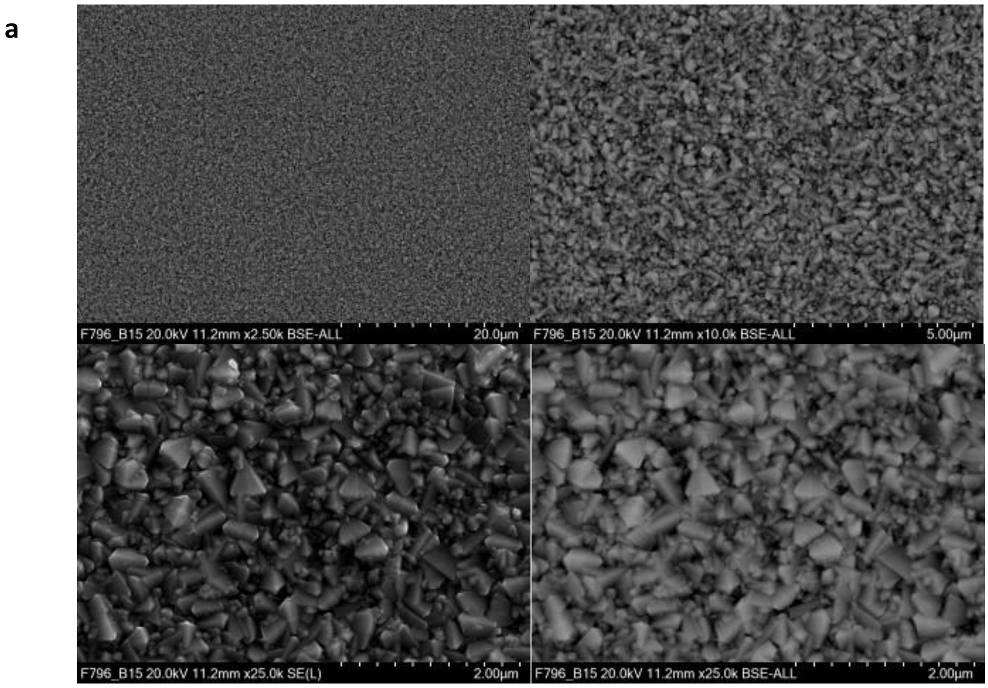
Supporting Information



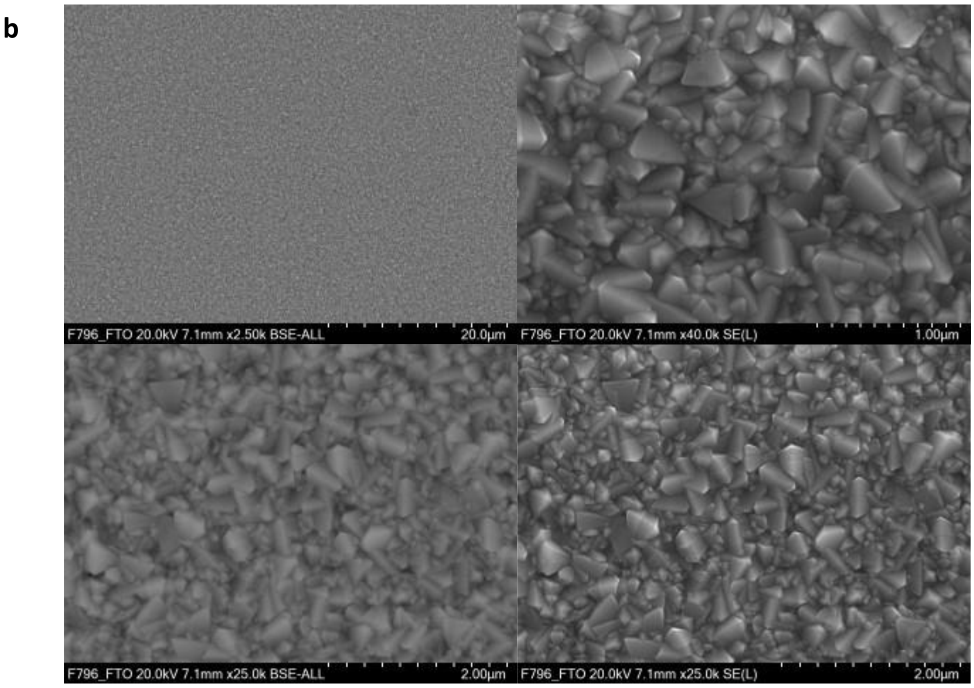


Figure S1: SEM images of an InSbO$_x$/CuSb$_2$O$_x$/FTO electrode (a) and an FTO substrate (b) at different magnifications.

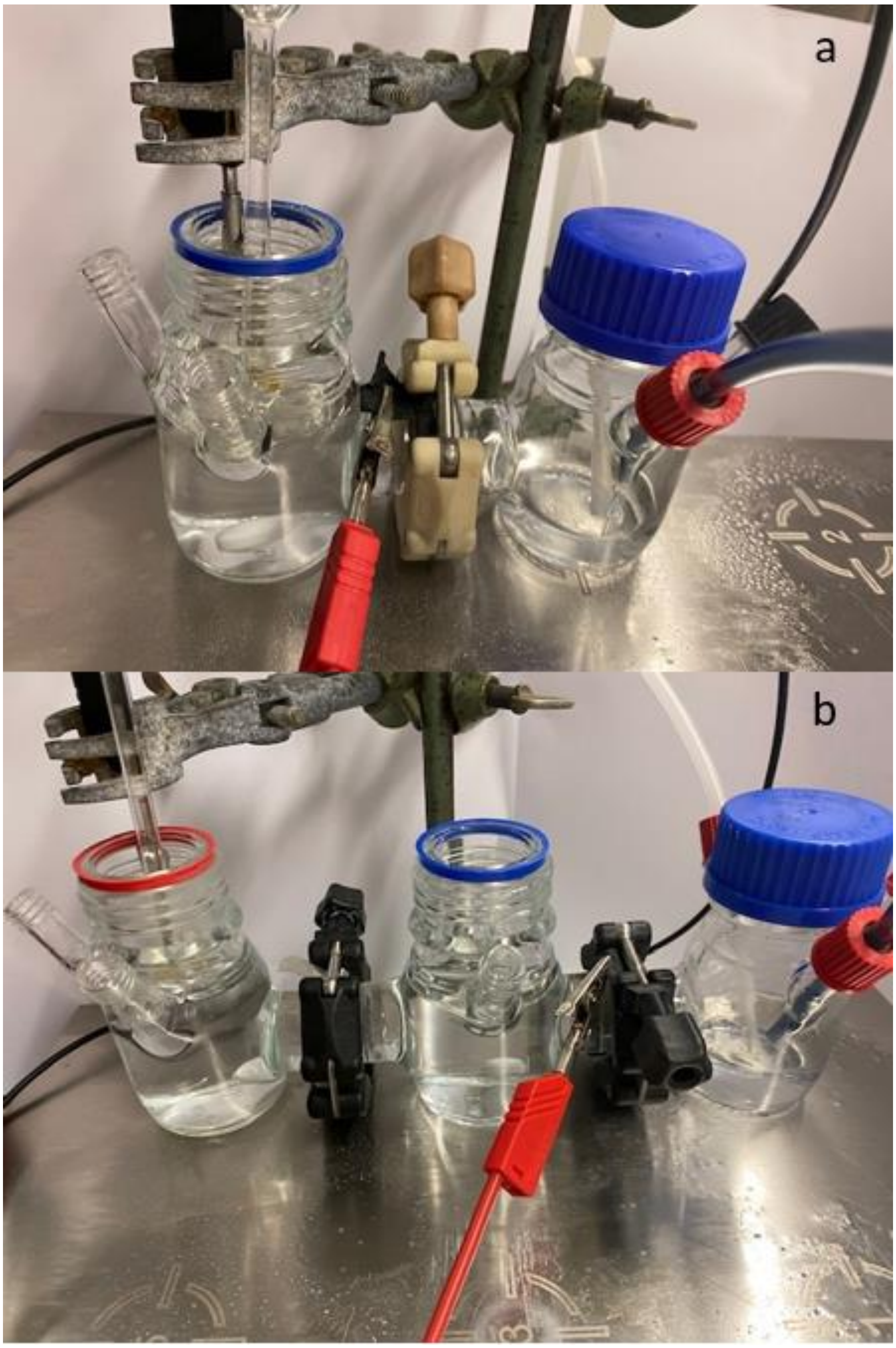


Figure S2: Photographs of the H-cells used. The single H-cell without membrane (a) and the double H-cell with membrane (b).

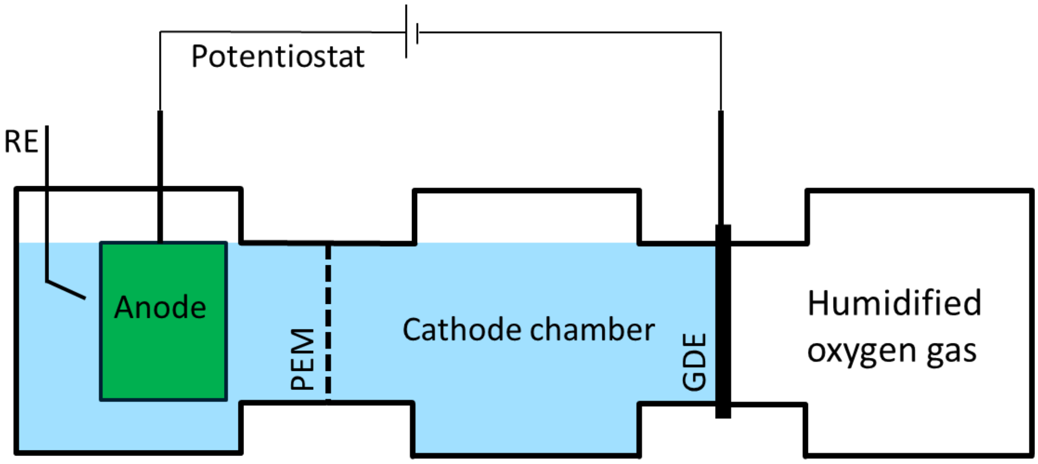


Figure S3: Illustration of the 3-chamber setup used for the experiments with a membrane.

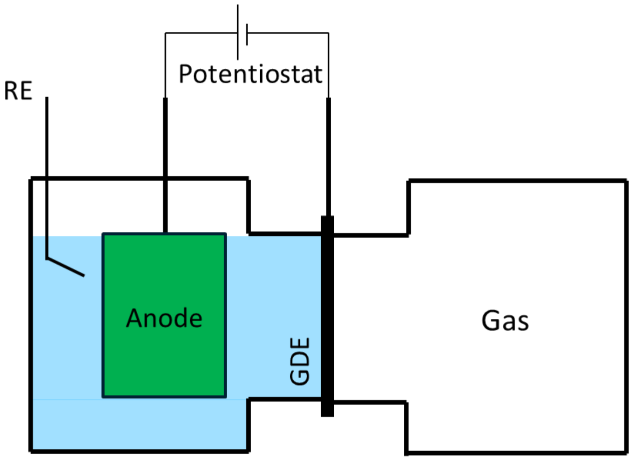


Figure S4: Illustration of the 2-chamber setup used for the experiments without a membrane.

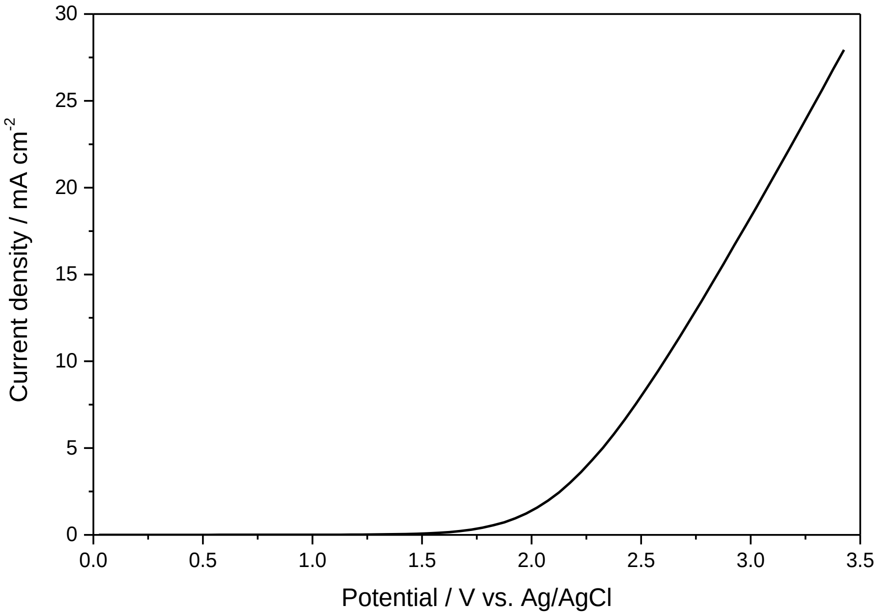


Figure S5: Linear sweep voltammogram of a $InSbO_x/CuSb_2O_x$/FTO as working electrode (2.4 cm^2). The measurement was performed in an unseparated cell using the GDE with carbon black as counter electrode (geometrical area 4.9 cm^2). Polarization was performed against Ag/AgCl/KCl(sat.) in carbonate buffer under stirring with 0.5 M $KHCO_3$ and 3.5 M K_2CO_3 at a pH of 11.3. The cell volume was 120 mL of electrolyte solution. In the gas chamber of the GDE, humidified oxygen was offered in the GDE with an overpressure of 15 mbar.

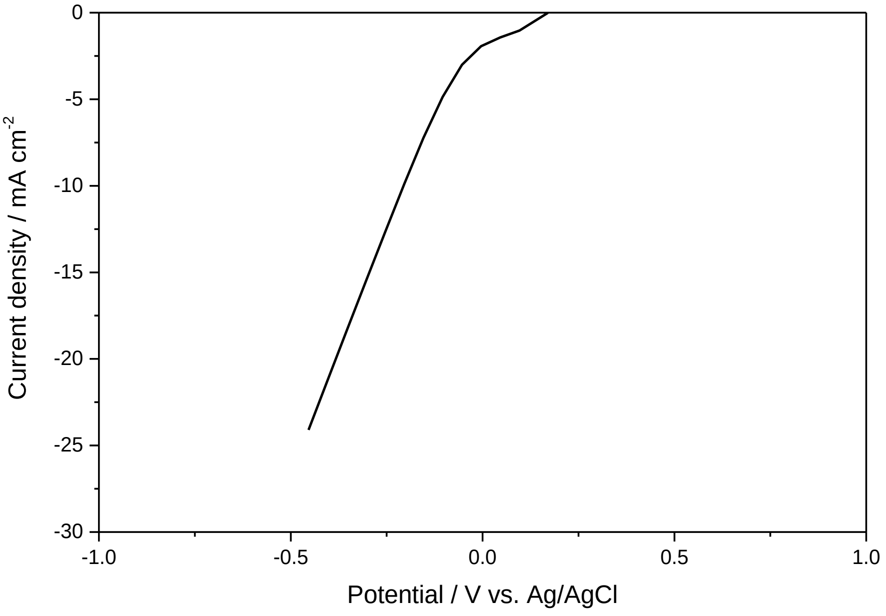


Figure S6: Linear sweep voltammogram of a GDE with carbon black as working electrode (4.9 cm^2). The measurement was performed in an unseparated cell using the $InSbO_x/CuSb_2O_x$/FTO electrode as counter electrode (geometrical area 2.4 cm^2). Polarization was performed against Ag/AgCl/KCl(sat.) in carbonate buffer under stirring with 0.5 M $KHCO_3$ and 3.5 M K_2CO_3 at a pH of 11.3. The cell volume was 120 mL of electrolyte solution. In the gas chamber of the GDE, humidified oxygen was offered in the GDE with an overpressure of 15 mbar.

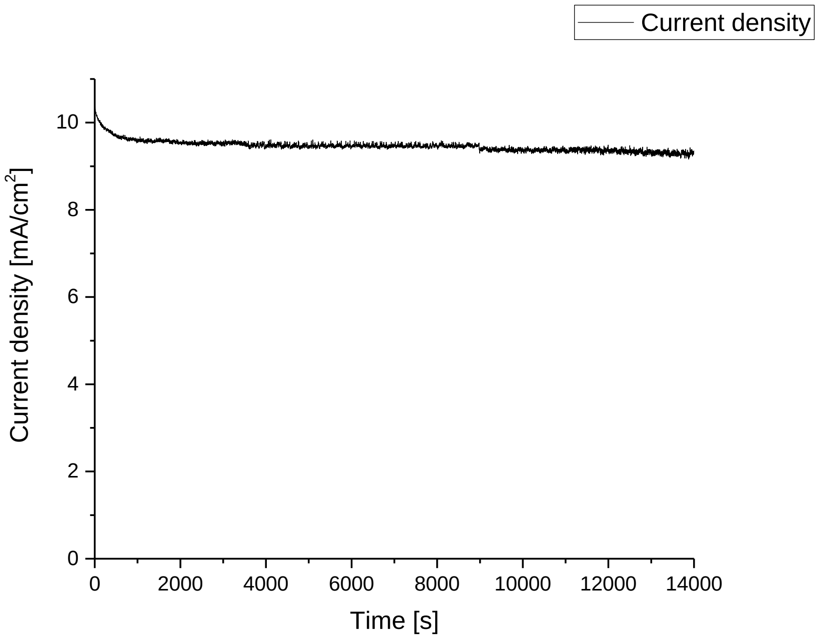


Figure S7: The current over time profiles of the chronocoulometry results depicted in Figure 3. Chronocoulometry of an $InSbO_x/CuSb_2O_x$/FTO electrode (2.4 cm^2) as anode. A GDE with carbon black (4.9 cm^2) was used as the cathode. Polarization was carried out at 2.385 V vs. Ag/AgCl (3.25 V vs. RHE) in carbonate buffer while stirring with 0.5 M $KHCO_3$ and 3.5 M K_2CO_3 at a pH of 11.3. The anode and cathode compartments each contained 120 mL of electrolyte and were separated from one another by a cation exchange membrane. In the gas room of the GDE, humidified oxygen was offered at 15 mbar overpressure.

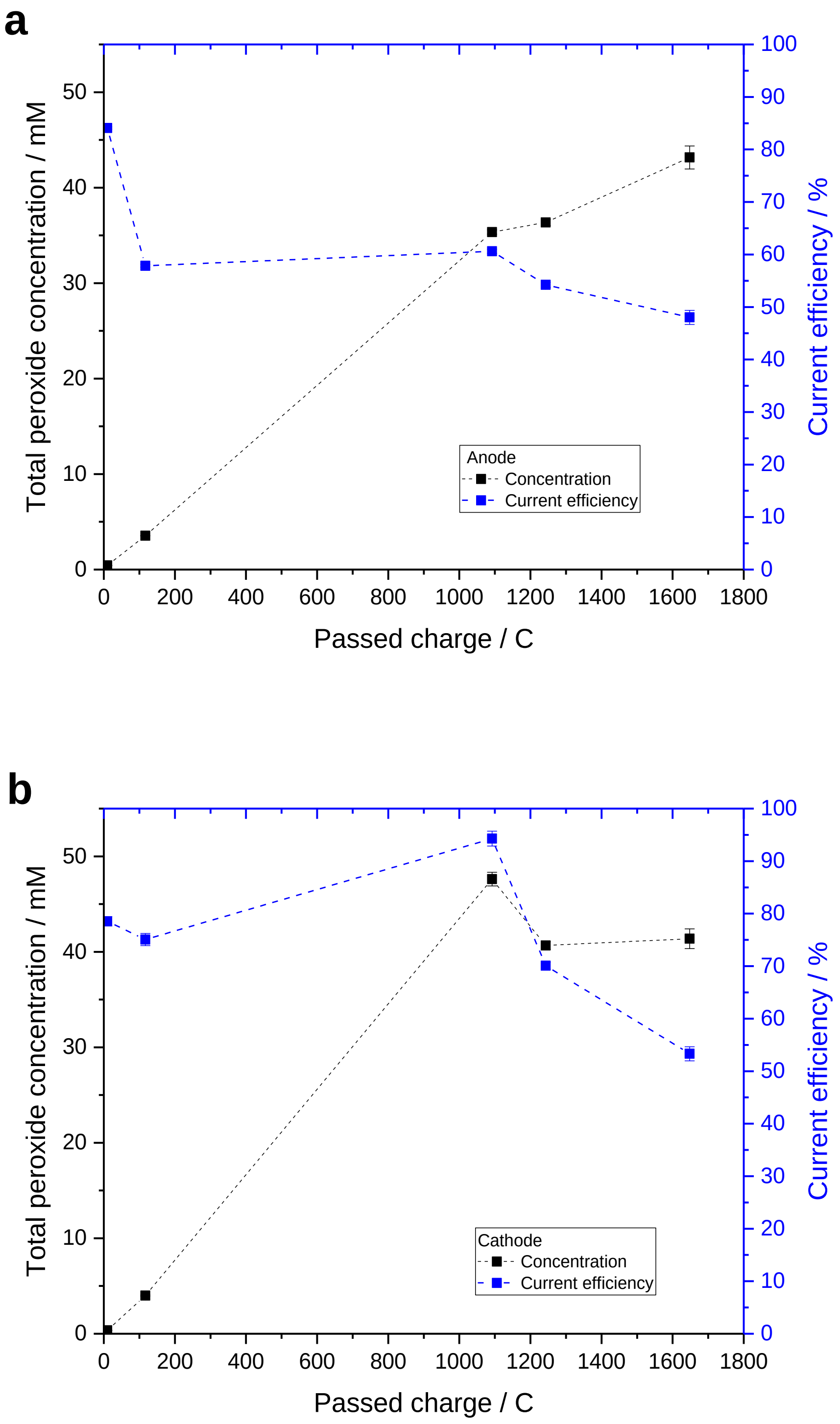


Figure S8: Repeated measurement for Figure 3. The total peroxide concentration and the CE plotted against the passed charge. (a) Peroxide production in the anode compartment of an InSbO$_x$/CuSb$_2$O$_x$/FTO electrode (2.4 cm^2). (b) Peroxide production in the cathode compartment of a GDE with carbon black (4.9 cm^{-2}). Chronocoulometry with InSbO$_x$/CuSb$_2$O$_x$/FTO as working electrode. Polarization was carried out at 2.385 V vs. Ag/AgCl (3.25 V vs. RHE) in stirred carbonate buffer with 0.5 M $KHCO_3$ and 3.5 M K_2CO_3 at a pH of 11.3. The anode and cathode compartments each contained 120 mL of electrolyte solution and were separated from one another by a cation exchange membrane. In the gas room of the GDE, humidified oxygen was offered at 15 mbar overpressure. Error bars indicate the results of repeated peroxide analysis (n=2-3) of this singular electrochemical experiment. The interpolated lines between the data points only serve as a visual guide.

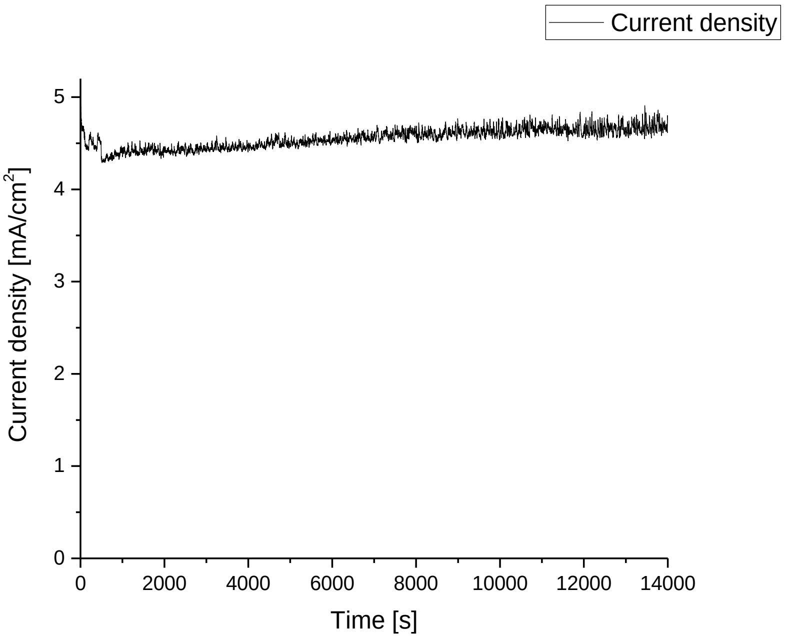


Figure S9: Chronocoulometry of a FTO substrate (2.3 cm^2) as anode. A GDE with carbon black (4.9 cm^2) was used as the cathode. Polarization was carried out at 2.385 V vs. Ag/AgCl (3.25 V vs. RHE) in carbonate buffer while stirring with 0.5 M $KHCO_3$ and 3.5 M K_2CO_3 at a pH of 11.3. The anode and cathode compartments each contained 120 mL of electrolyte and were separated from one another by a cation exchange membrane. In the gas room of the GDE, humidified oxygen was offered at 15 mbar overpressure.

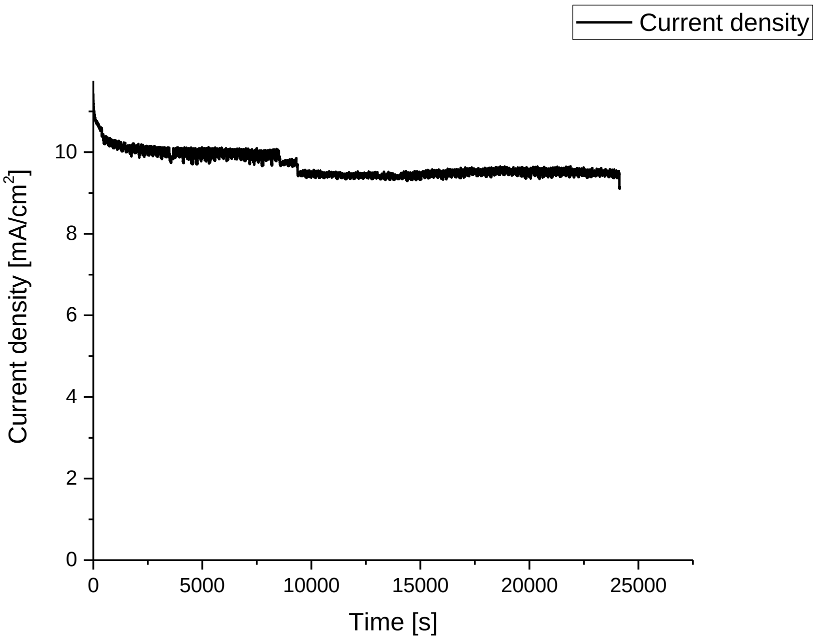


Figure S10: Chronoamperometry of an $InSbO_x/CuSb_2O_x$/FTO electrode (2.4 cm^2) as anode. A GDE with carbon black (4.9 cm^2) was used as the cathode. Polarization was carried out at 2.385 V vs. Ag/AgCl (3.25 V vs. RHE) in carbonate buffer while stirring with 0.5 M $KHCO_3$ and 3.5 M K_2CO_3 at a pH of 11.3 in an undivided cell. The electrode compartment contained 120 mL of electrolyte solution. In the gas room of the GDE, humidified oxygen was offered at 15 mbar overpressure.

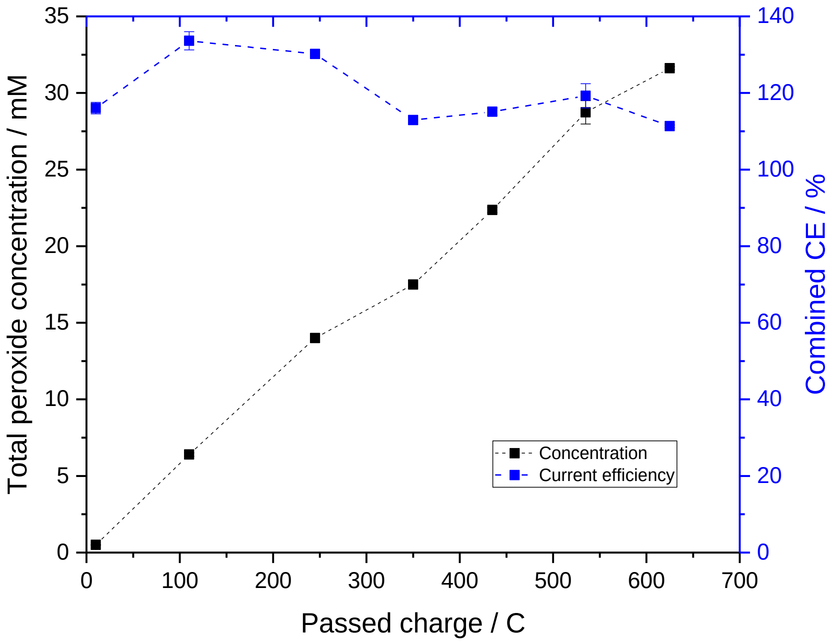


Figure S11: Repeated measurement for Figure 4. Total formed peroxide over passed charge, using an $InSbO_x/CuSb_2O_x$/FTO electrode (2.4 cm^2) as anode. A GDE with carbon black (4.9 cm^{-2}) was used as the cathode. Polarization was carried out at 2.385 V vs. Ag/AgCl (3.25 V vs. RHE) with the anode as working electrode in carbonate buffer while stirring with 0.5 M $KHCO_3$ and 3.5 M K_2CO_3 at a pH of 11.3. The undivided electrode compartment contained 120 mL of electrolyte solution. In the gas room of the GDE, humidified oxygen was offered at 15 mbar overpressure. Error bars indicate the results of repeated peroxide analysis (n=2-3) of this singular electrochemical experiment. The interpolated lines between the data points only serve as a visual guide.

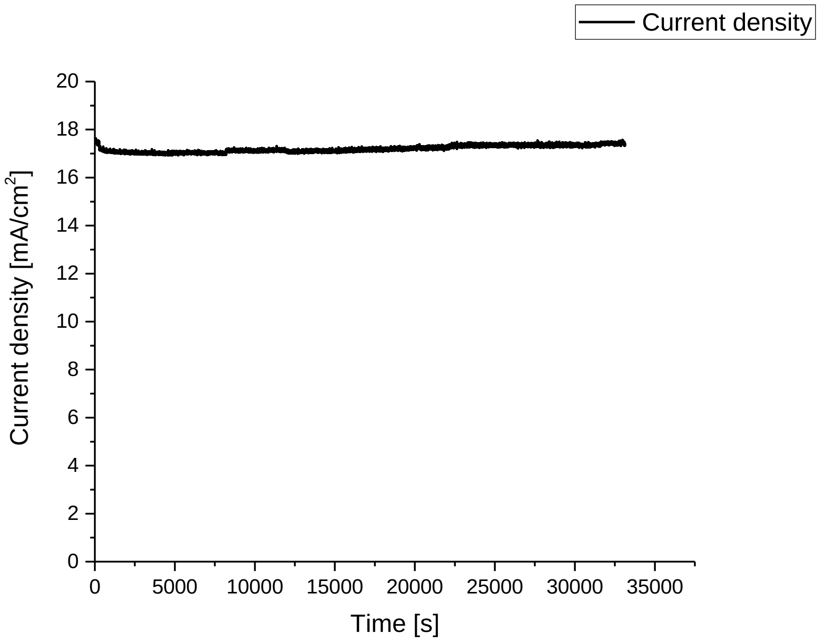


Figure S12: Chronoamperometry of an $InSbO_x/CuSb_2O_x$/FTO electrode (2.4 cm^2) as anode. A GDE with carbon black (4.9 cm^2) was used as the cathode. Polarization was carried out at 3 V vs. Ag/AgCl (3.85 V vs. RHE) in carbonate buffer while stirring with 0.5 M $KHCO_3$ and 3.5 M K_2CO_3 at a pH of 11.3. The electrode compartment contained 100 mL of electrolyte solution. In the gas room of the GDE, humidified oxygen was offered at 15 mbar overpressure.

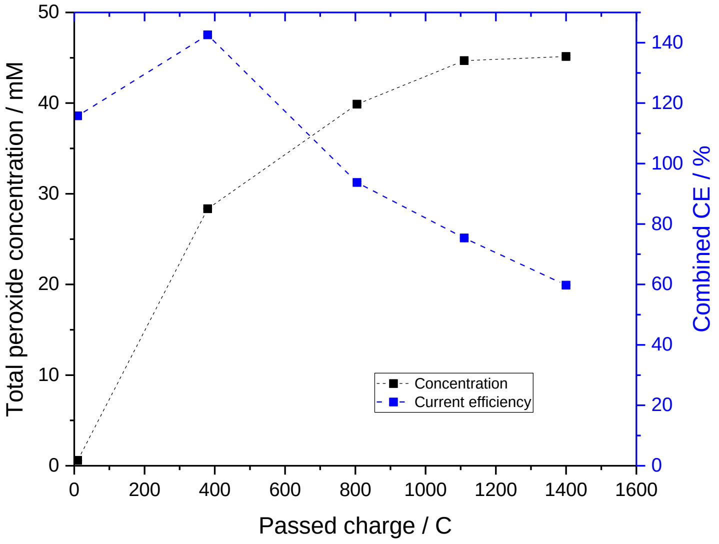


Figure S13: Repeated measurement for Figure 5. Total formed peroxide over passed charge, using an $InSbO_x/CuSb_2O_x$/FTO electrode (2.7 cm^2) as anode. A GDE with carbon black (4.9 cm^2) was used as the cathode. Polarization was carried out at 3 V vs. Ag/AgCl (3.85 V vs. RHE) in stirred carbonate buffer with 0.5 M $KHCO_3$ and 3.5 M K_2CO_3 at a pH of 11.3. The electrode compartment contained 100 mL of electrolyte solution. In the gas room of the GDE, humidified oxygen was offered at 15 mbar overpressure. Error bars indicate the results of repeated peroxide analysis (n=2-3) of this singular electrochemical experiment. The interpolated lines between the data points only serve as a visual guide.

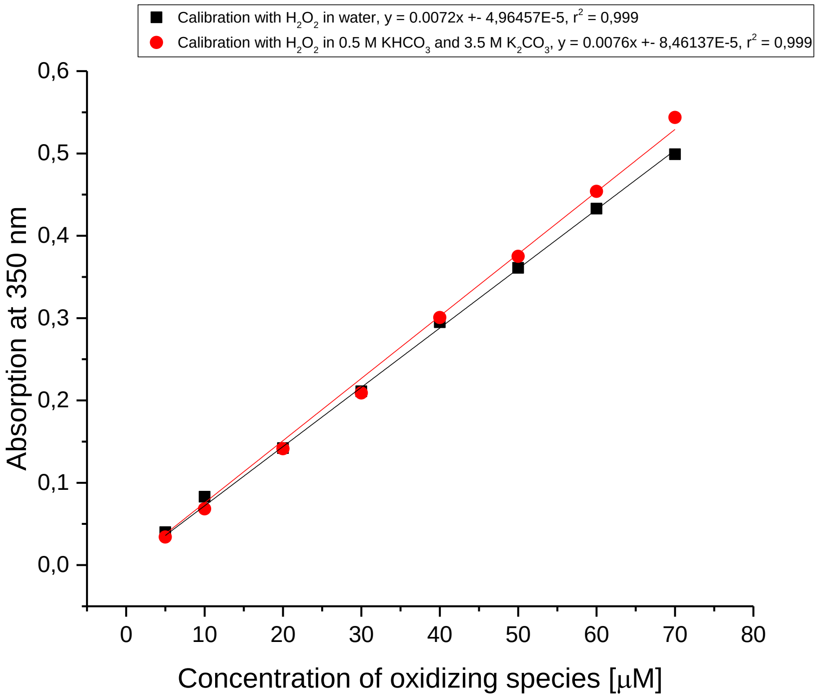


Figure S14: Calibration curves of the quantification assay based on colorimetric iodometry using either a fixed concentration of H_2O_2 in water (black) or in 0.5 M $KHCO_3$ and 3.5 M K_2CO_3 (red).

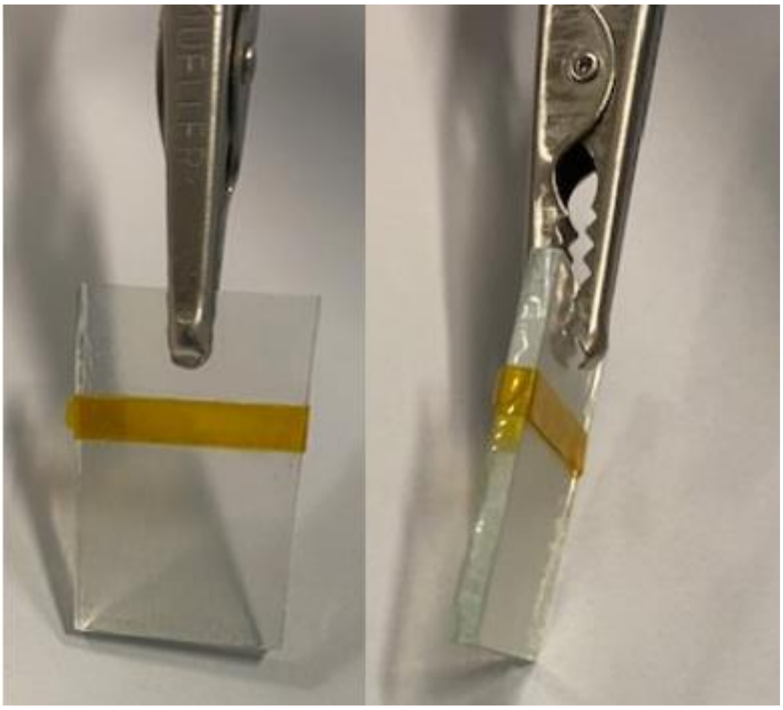


Figure S15: Contacting of the anode with a crocodile clip. Here you can see the front and the side view. The electrode is masked with kapton tape so that only a defined area comes into contact with the electrolyte.