# Efficiency and selectivity $CO_2$ reduction to CO on gold gas diffusion electrodes in acidic media

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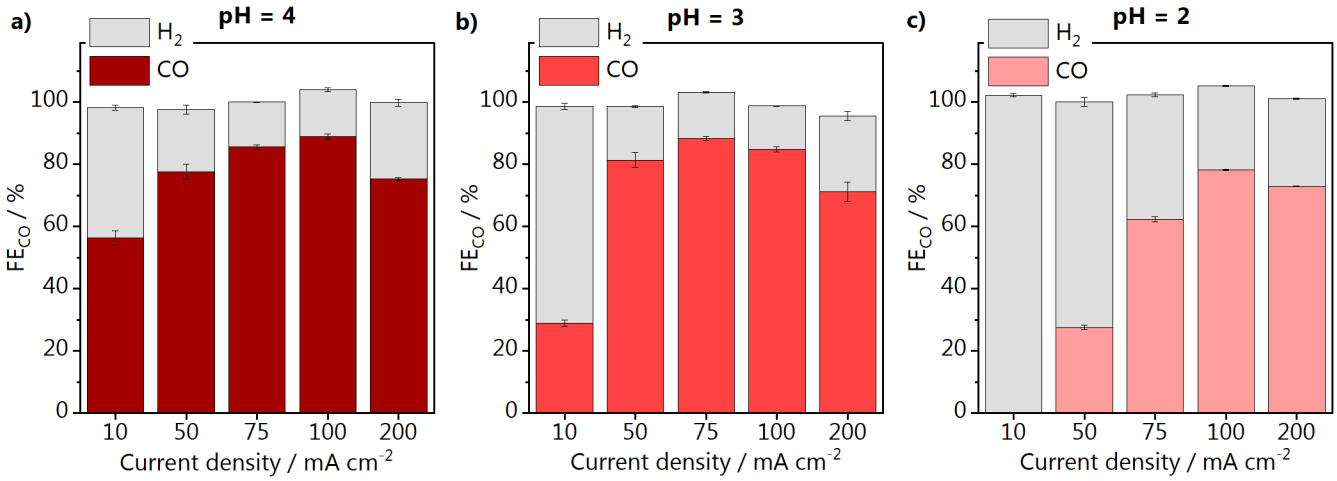
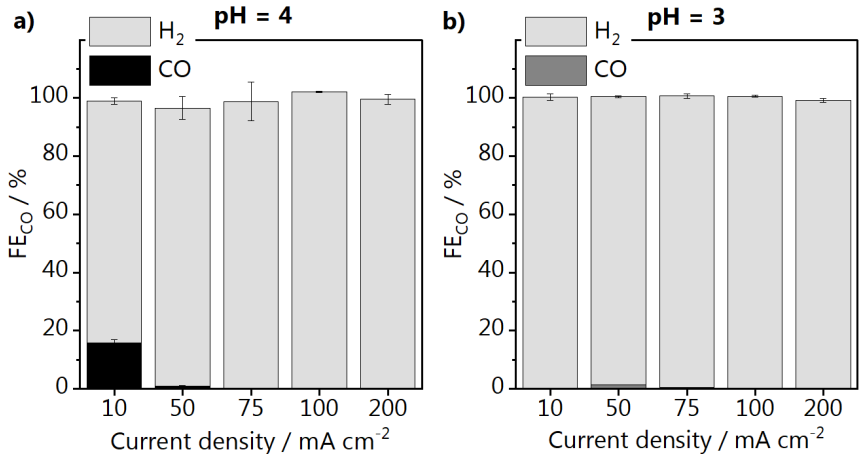
## Supplementary Information

## 1. Effect of pH



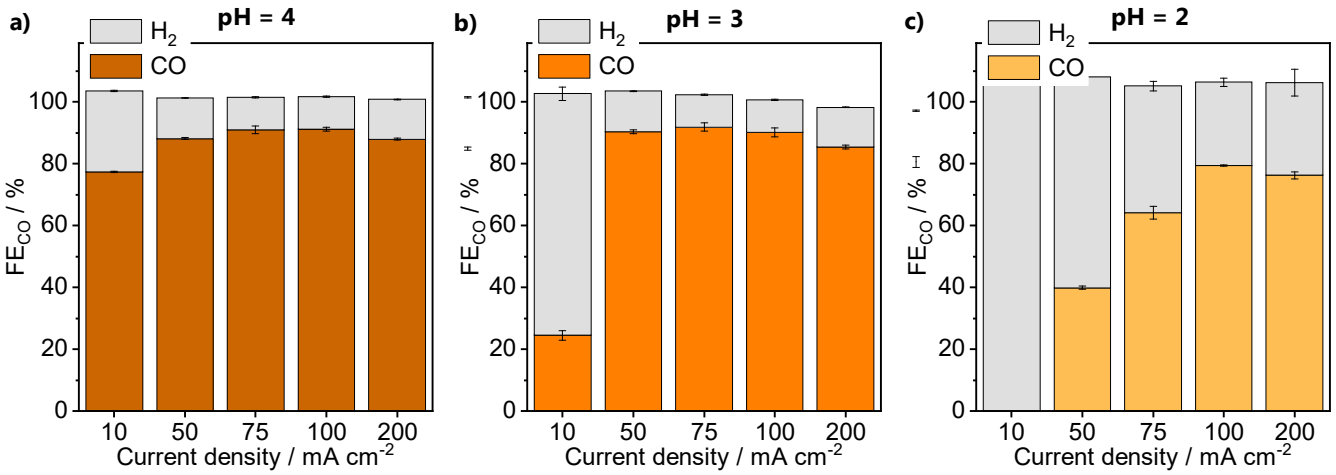

**Supplementary Figure 1.** Faradaic efficiencies for CO and $H_2$ obtained during one hour electrolysis at each current density displayed. The catholyte was 1 M $Cs_2SO_4$, **a)** pH 4, **b)** pH 3 and **c)** pH 2. Catalyst loading 2 mg $cm^{-2}$. Error bars are calculated based on three individual measurements.

## 2. Cation identity

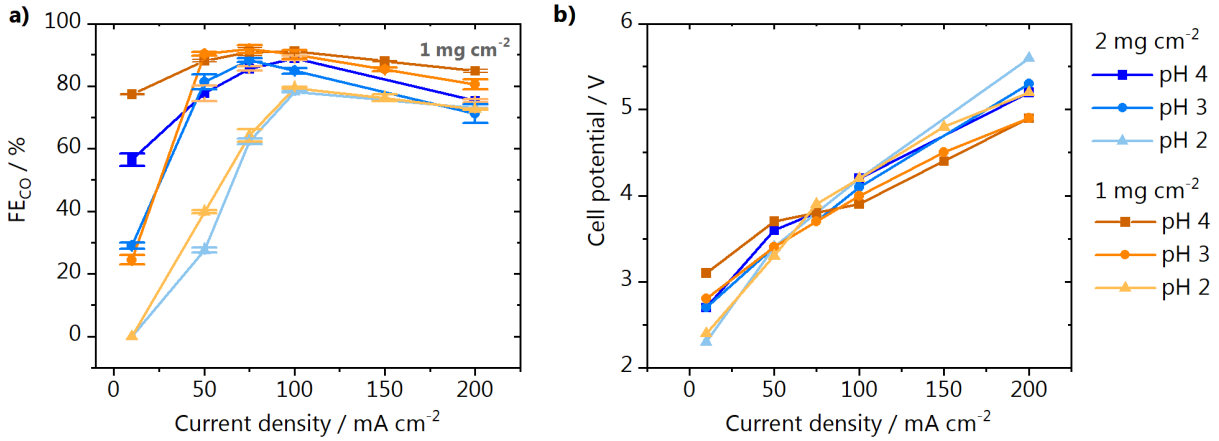



**Supplementary Figure 2.** Faradaic efficiencies for CO and $H_2$ obtained during one hour electrolysis at each current density displayed. The catholyte was 1 M $Li_2SO_4$, **a)** pH 4 and **b)** pH 3. Catalyst loading 2 mg $cm^{-2}$. Error bars are calculated based on three individual measurements.

## 3. Effect of the catalyst loading

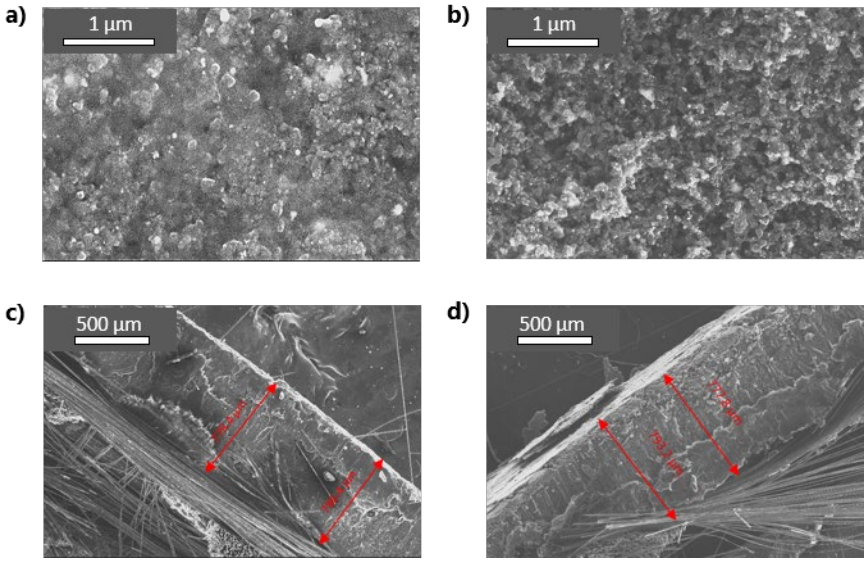



**Supplementary Figure 3.** Faradaic efficiencies for CO and $H_2$ obtained during one hour electrolysis at each current density displayed. The catholyte was 1 M $Cs_2SO_4$; **a)** pH 4, **b)** pH 3 and **c)** pH 2. Catalyst loading 1 mg $cm^{-2}$. Error bars are calculated based on three individual measurements.

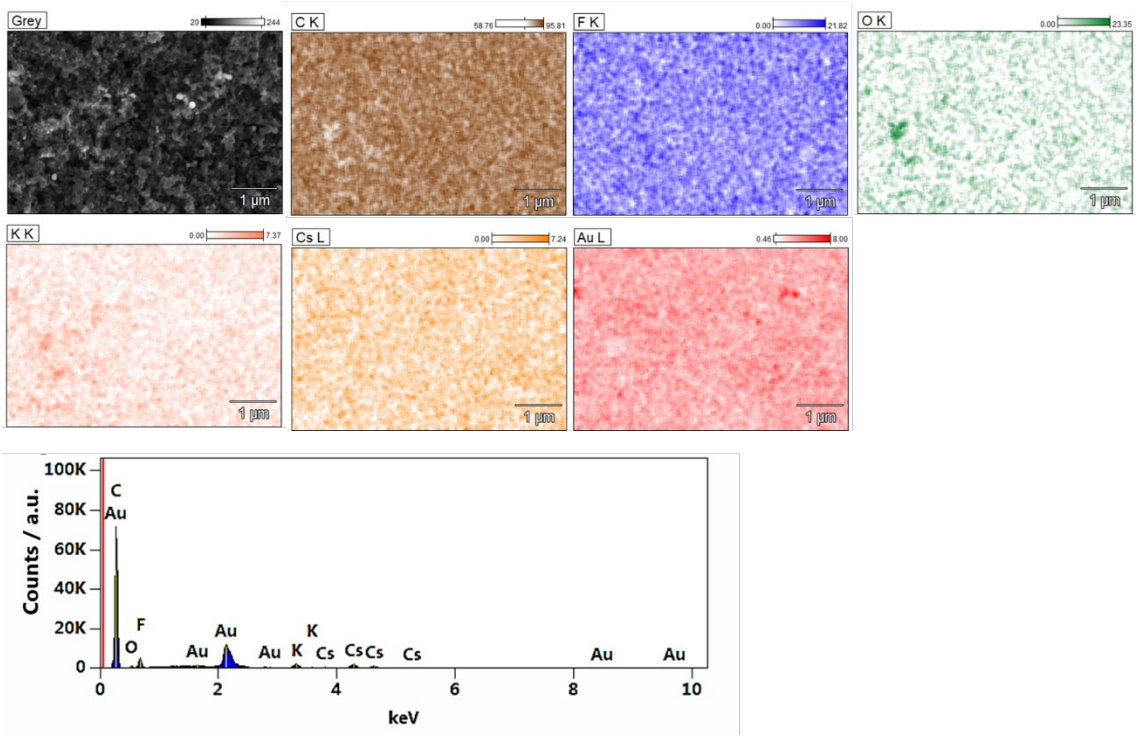



**Supplementary Figure 4.** Electrolysis performed in 1 M $Cs_2SO_4$, catalyst loading 1 mg $cm^{-2}$ or 2 mg $cm^{-2}$, **a)** faradaic efficiency for CO and **b)** cell potential. The data points displayed are an average obtained during 1 hour electrolysis. Error bars are calculated based on three individual measurements.

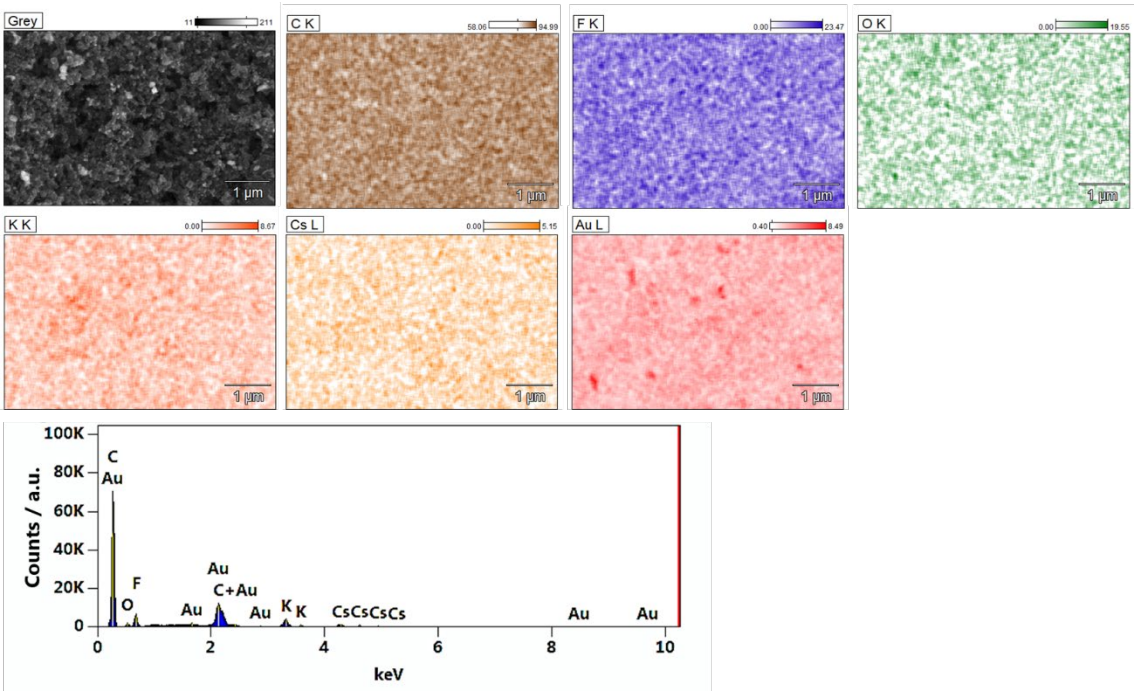
## 4. SEM micrographs and EDX elemental analysis of the GDEs with different loadings.




**Supplementary Figure 5.** SEM micrograph of the gold GDEs. Top view of the **a)** 1 mg cm$^{-2}$ and **b)** 2 mg cm$^{-2}$ catalyst, and cross-section of the **c)** 1 mg cm$^{-2}$ and **d)** 2 mg cm$^{-2}$ catalyst.

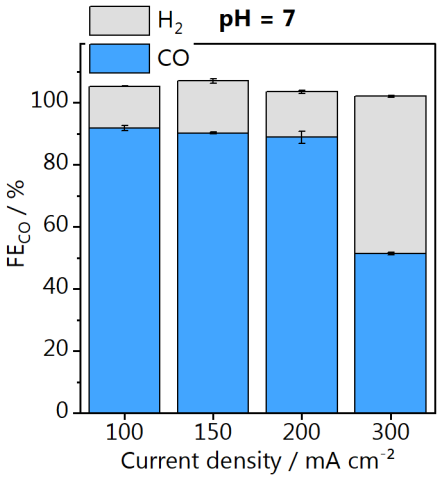



**Supplementary Figure 6.** SEM micrograph, EDX elemental map and spectrum of the gold GDE with 1 mg cm$^{-2}$ loading.

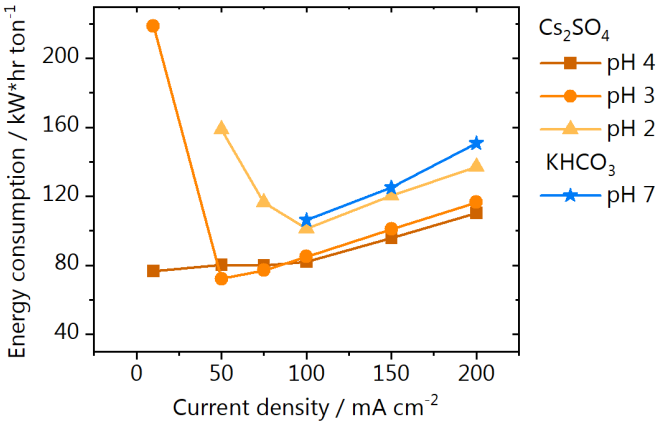



**Supplementary Figure 7.** SEM micrograph, EDX elemental map and spectrum of the gold GDE with 2 mg $cm^{-2}$ loading.

## 5. Comparison with bicarbonate




**Supplementary Figure 8.** Faradaic efficiencies for CO and $H_2$ obtained during one hour electrolysis at each current density displayed. The catholyte was 1 M $KHCO_3$ (pH 7) and the catalyst loading 1 mg $cm^{-2}$. Error bars are calculated based on three individual measurements.




**Supplementary Figure 9.** Energy consumption per ton of CO produced, based on the faradaic efficiencies and cell potentials obtained for electrolysis in 1 M $Cs_2SO_4$ or 1 M $KHCO_3$ at different current densities (catalyst loading 2 mg $cm^{-2}$).

Energy efficiency calculation:

$$\varepsilon_{energy} = \frac{\left(E^0_{CO_2} + E^0_{O_2}\right) * FE_{CO}}{E_{Cell}} \qquad \text{Supplementary Equation 1}$$

where $E^0$ are the standard potentials of the cathode and anode reactions, *FE* is the faradaic efficiency as percentage, and *Ecell* is the cell potential in V.

Energy consumption calculation:

$$\frac{kW*h}{ton} = \frac{E_{cell}*n*F}{MW*FE_{CO}*3.6} \qquad \text{Supplementary Equation 2}$$

where *n* is the number of electrons transferred (dimensionless), *F* is Faraday´s constant in C $mol^{-1}$, *MW* is the molecular weight of CO in g $mol^{-1}$.

## 6. Comparison with literature

Supplementary Table 1. Experimental details of the different work from literature, used for the comparison made in Figure 5 in the main text. Only GDEs with geometrical surface area equal or larger than 5 cm² were considered.

| | Cathode (GDE) | Anode | Electrolyte | $CO_2$ flow (mL $min^{-1}$) | Load (mg $cm^{-2}$) | Cathode area ($cm^2$) | Ref |
|---|---|---|---|---|---|---|---|
| this work | Au nanoparticles | DSA® | 1 M $Cs_2SO_4$ | 50 | 1 | 10 | this work |
| Haas 2018 | Ag GDE Covestro | Ir-MMO/Ti sheet | 0.1 M $K_2SO_4$ + 1.5 M $KHCO_3$ | 52 and 10.5 | n.a. | 10 | [1] |
| Verma 2018 | MWNT/PyPBI/Au on Sigracet® 35 BC | $IrO_2$ | 2 M $KHCO_3$ or 2 M KOH | 17 | 1 | 5 | [2] |
| Verma 2016 | Ag NP on Sigracet® 35 BC | $IrO_2$ | 1 M $KHCO_3$ | 17 | 2 | 10 | [3] |
| Duarte 2019 | Ag NP on Sigracet® 39 BC | platinized Ti | 2 M $KHCO_3$ | 77 | 0.75 | 10 | [4] |
| Jeanty 2018 | Ag GDE Covestro | n.a. | 0.4 M $K_2SO_4$ | 21 | n.a. | 7.67 | [5] |

## Supplementary References


1. Haas, T., Krause, R., Weber, R., Demler, M. & Schmid, G. Technical photosynthesis involving $CO_2$ electrolysis and fermentation. *Nat. Catal.* **1**, 32–39 (2018).
2. Verma, S. *et al.* Insights into the Low Overpotential Electroreduction of $CO_2$ to CO on a Supported Gold Catalyst in an Alkaline Flow Electrolyzer. *ACS Energy Lett.* **3**, 193–198 (2018).
3. Verma, S., Lu, X., Ma, S., Masel, R. I. & Kenis, P. J. A. The effect of electrolyte composition on the electroreduction of $CO_2$ to CO on Ag based gas diffusion electrodes. *Phys. Chem. Chem. Phys.* **18**, 7075–7084 (2016).
4. Duarte, M., De Mot, B., Hereijgers, J. & Breugelmans, T. Electrochemical Reduction of $CO_2$: Effect of Convective $CO_2$ Supply in Gas Diffusion Electrodes. *ChemElectroChem* **6**, 5596–5602 (2019).
5. Jeanty, P. *et al.* Upscaling and continuous operation of electrochemical $CO_2$ to CO conversion in aqueous solutions on silver gas diffusion electrodes. *J. CO2 Util.* **24**, 454–462 (2018).