VP 10

Microwave assisted synthesis of metal organic chalcogenolate assemblies as electrocatalysts for syngas production

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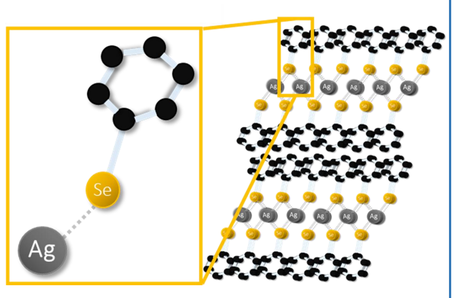
1. Introduction

1.1 Motivation

Electrocatalytic CO_2 reduction in aqueous media is an efficient method for reducing the CO_2 in our atmosphere and converting it into a more valuable feedstock such as for example syngas, a mixture of H_2 and CO. It further has the great advantage of being operated at room temperatures and ambient pressures. One major challenge in the field however is the search for selective, stable and efficient catalysts.

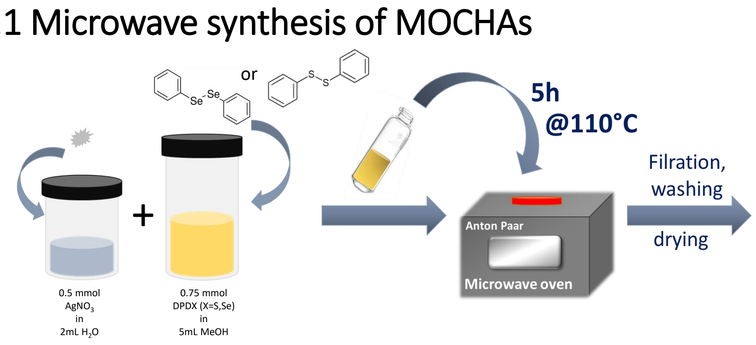
1.2 Metal organic chalcogenolate assemblies (MOCHAs)

Metal organic chalcogenolate assemblies (MOCHAs) such as $[AgSePh]_\infty$ also referred to as mithrene or $[AgSPh]_\infty$ (thiorene) are organic-inorganic hybrid materials with outstanding optoelectronic properties[1,2]. They were recently reported to work as X-ray[3] and Pb^{2+} sensors[4]. This work has shown their potential as suitable electrocatalysts for syngas formation[5].

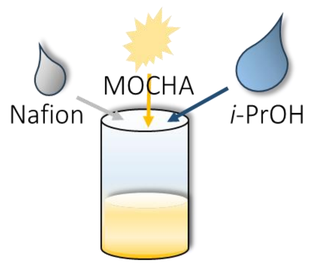


2. Synthesis

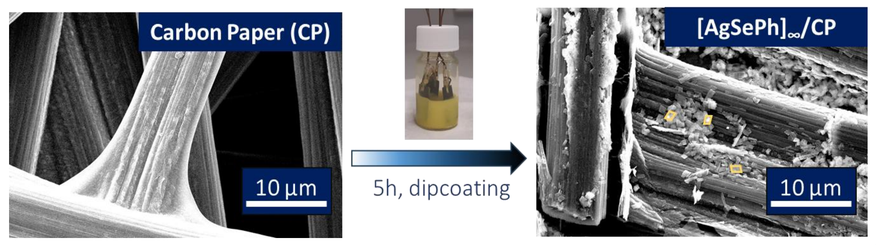
2.1 Microwave synthesis of MOCHAs



2.2 Ink preparation

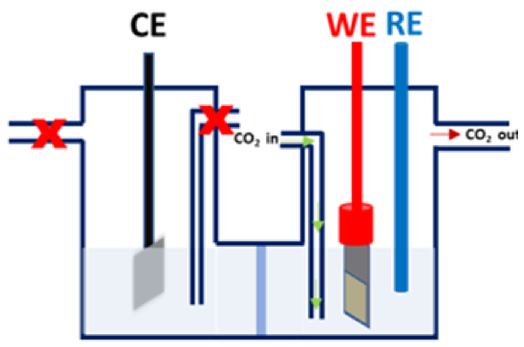


2.3 Dipcoating process



3. Electrocatalytic Syngas formation

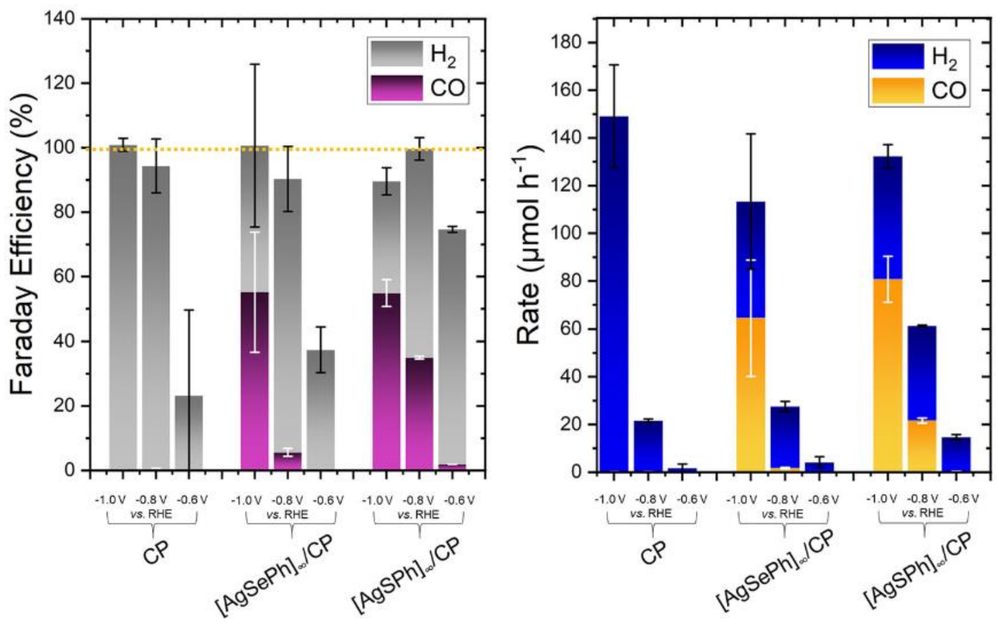
3.1 H-cell setups



An H-cell setup filled with 0.5M $KHCO_3$ on both sides was chosen. The working side compartment was saturated with CO_2 prior to electrolysis. The MOCHA/CP electrodes were used as working electrodes. An Ag/AgCl/3M KCl was used as reference electrode and Pt-foil was used as counter electrode.

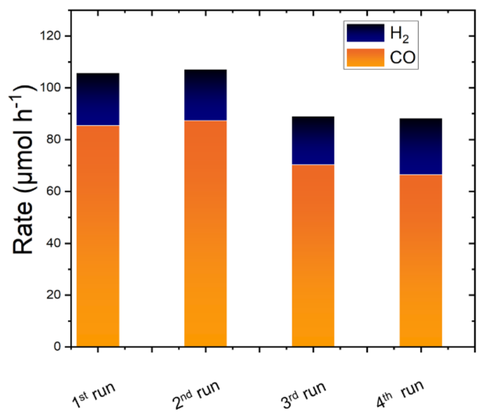
3.2. Syngas formation

Bare CP does not lead to any CO production. Thiorene ($[AgSPh]_\infty$) outperforms mithrene ($[AgSePh]_\infty$) by approx. 17 µmol h^{-1} regarding CO production. The syngas ratio (H_2:CO) can be tuned from 1 to 10 depending on the applied potential.



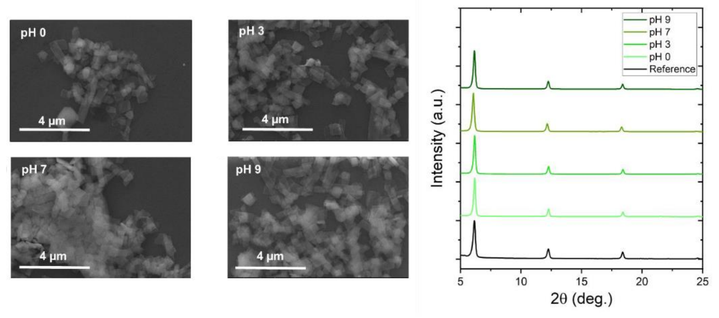
4. Stability studies

4.1 Recyclability of $[AgSePh]_\infty$ upon electrolysis

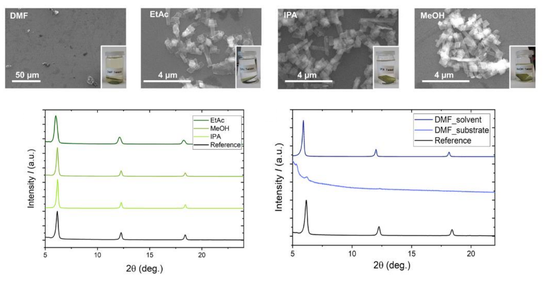


Using the same $[AgSePh]_\infty$/CP electrode for four consecutive electrolysis runs revealed recyclability of the MOCHA catalyst. A stable CO:H_2 ratio was obtained for all four runs.

4.2 Stability of $[AgSePh]_\infty$ upon various conditions

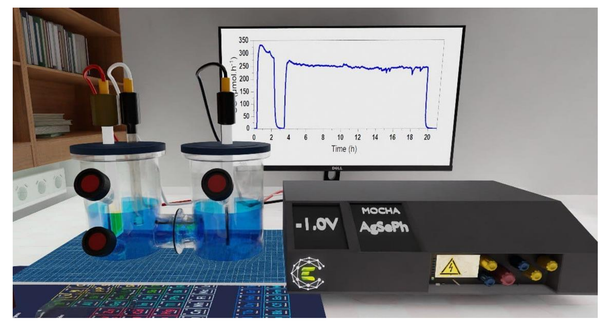


Stability tests reveal structural (pXRD) and morphological (SEM) stability of $[AgSePh]_\infty$ under prolonged exposure (>1 week) to a wide pH range and various organic solvents.



6. Take-home messages

- A facile, fast and upscalable synthesis method for metal organic chalcogenolate assemblies was developed.
- First application of the MOCHAs $[AgSePh]_\infty$ (mithrene) and $[AgSPh]_\infty$ (thiorene) for electrocatlytic syngas formation was shown.
- The ratio of syngas (CO:H_2) can be tuned by the applied potential.
- Recyclability and stability of $[AgSePh]_\infty$ was shown upon 4 consecutive electrolysis runs.
- MOCHAs are stable over a wide pH and temperature range and in various organic solvents.



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References

[1] Schriber, E. A. *et al.* Mithrene Is a Self-Assembling Robustly Blue Luminescent Metal–Organic Chalcogenolate Assembly for 2D Optoelectronic Applications. *ACS Appl. Nano Mater.* **1**, 3498–3508 (2018).

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Acknowledgements

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