

High Current Density Electrochemical Reduction of CO₂ to Formate using Tin-loaded Gas Diffusion Electrodes

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Background

The electrochemical utilization of CO₂ using electricity from renewable resources is a promising way of reducing CO₂ emissions in existing chemical processes. Besides that, CO₂ can be used as feedstock for producing chemicals or as energy storage chemical. Formic acid or formate is one possible product for these tasks, since conventional methods produce formic acid with poor energy and atom efficiency. Formic acid can easily be decomposed releasing hydrogen for conventional fuel cells, or as formate solution, which can be used in a direct formate fuel cell, making it a perfectly safe and liquid energy storage, not only in industrial but also for mobile applications. [1] Although this topic experienced a lot of interest in recent years, questions for a technical realization remain unanswered.

Challenges

The main objective of this project is to improve the process efficiency by optimizing the process parameters and reactor design, as well as by enhancing the performance of the nano-sized catalyst, the used gas diffusion electrode (GDE) and electrolyte. Important characteristics like faradaic and energetic efficiency or current density have thereby to be maximized. In order to avoid mass transport limitations, caused by the low solubility of CO₂ in aqueous media, porous GDEs are used, in which the electrocatalyst is dispersed. GDEs provide a huge internal surface and can widely enhance the three-phase boundary that is necessary for the aqueous CO₂ reduction.

Results

The enhanced performance of tin-loaded GDE's has already been shown by our group with current densities of up to 400 mA cm⁻² at a faradaic efficiency (selectivity) of about 90 % [2]. In addition, the general technical feasibility in continuous mode of operation has already been investigated and confirmed [3]. Recently, we focused on

the temperature as an important process parameter and achieved outstanding results with supported tin-oxide nanoparticles of about 10 nm in diameter, on carbon (Acetylene Black), with regard to current density.

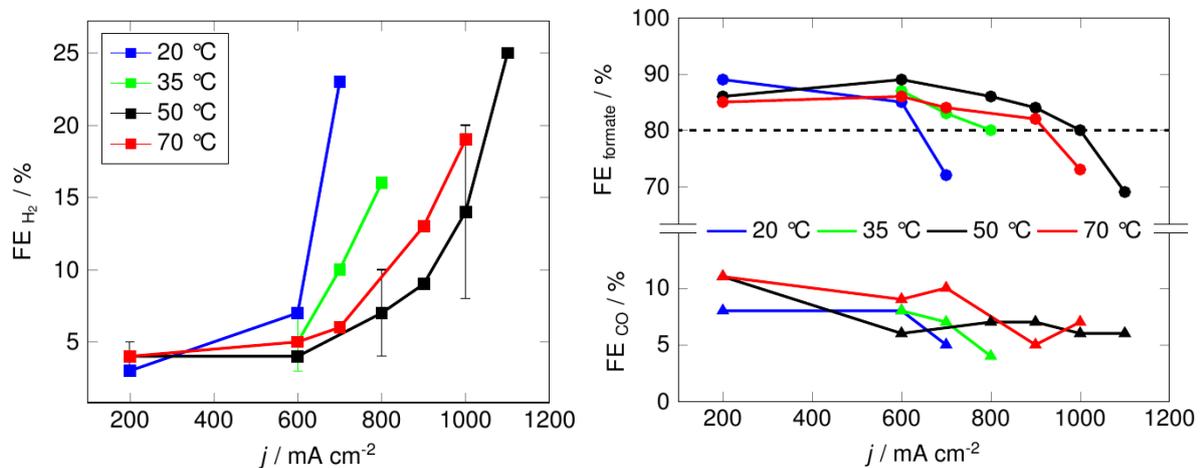


Figure 1: Faradaic efficiency towards H₂ (left) and formate (circle) and CO (triangle) (right), as a function of the current density at different temperatures. Values are averaged over 40 min of galvanostatic scan.

By keeping the temperature stable, the maximum current density with a minimum faradaic efficiency to formate of 80 % was shifted from 600 mA cm⁻² at 20 °C to 1000 mA cm⁻² at an optimum temperature of 50 °C. Up to this point, the positive effects of an increased electrode wetting and enhanced kinetics predominate the loss of CO₂-solubility. Above 50 °C, mass transport limitation again impairs the performance. In order to reach such high current densities, the electrolyte has to be suitable. Current research focuses on optimum conductivity and electrode wetting with simultaneous increased CO₂-solubility and availability as well as a favored formate production and minimum operating costs. Another very important part for an energy efficient production is the catalysts activity. Currently we are focusing on industrial reasonable solutions to enhance the performance, like an enhanced dispersion and accessibility, as well as on alloys and additives.

Literature

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- [3] D. Kopljar, N. Wagner, E. Klemm, *Chem. Eng. Technol.*, 39 (2016) p. 855 - 859