

Cover Page



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Title: Electrocatalytic carbon dioxide reduction : a mechanistic study

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Propositions

accompanying the thesis

Electrocatalytic carbon dioxide reduction

A mechanistic study

1. Using renewable electricity to replace electricity produced by carbon-intensive energy sources would likely result in a greater reduction in emissions than using that electricity to convert carbon dioxide. Still, the electrochemical conversion of carbon dioxide has great potential [...] because it provides a means of storing renewable electricity in a convenient, high-energy-density form.

D. T. Whipple and P. J. A. Kenis, J. Phys. Chem. Lett., 2010, 1, 3451-3458

2. The general reaction pathway from carbon dioxide to hydrocarbons is via the intermediates carboxyl and carbon monoxide.

Chapter 2 of this thesis

3. In contradiction to the outcome of theoretical calculations, formaldehyde is not an intermediate in the electrochemical reduction of carbon dioxide

Chapter 3 of this thesis

A. A. Peterson, F. Abild-Pedersen, F. Studt, J. Rossmeisl and J. K. Nørskov, Energy Environ. Sci., 2010, 3, 1311-1315

4. Rather than a Fischer-Tropsch like mechanism, a reaction mechanism via an enediol(ate) or oxametallacycle intermediate explains the selective ethylene formation during the electrochemical reduction of carbon dioxide.

Chapters 3 and 6 of this thesis.

5. There are at least two different reaction pathways towards the formation of ethylene during the electrochemical reduction of carbon dioxide on copper electrodes.

Chapter 5 of this thesis

6. The lower overpotential observed for the C-C bond formation in ethylene on Cu(100) during the electrochemical carbon dioxide reduction is not exceptional; other electrocatalytic reactions involving C-C, C-O, N-O and N-N bond breaking/making also prefer the (100) site geometry.

M. T. M. Koper, Nanoscale, 2011, 3, 2054-2073

7. In water-based electrolytes, hydrogen suppression during carbon dioxide reduction is challenging. It would be better to focus on an optimal efficiency for both hydrogen and carbon monoxide (syngas) or hydrogen and hydrocarbons (*e.g.* 'hythane').
8. The need to reduce carbon dioxide while avoiding high energy intermediates (*i.e.* reduce the required overpotential) requires a strong interplay between theoretical and experimental research, so that the understanding gained from one approach can complement the other.
9. Christianity, if false, is of no importance, and if true, of infinite importance. The only thing it cannot be is moderately important.

C. S. Lewis

Klaas Jan Schouten
Leiden, October 8, 2013