The role of CO* as a spectator in CO2 electro-reduction on RuO2 - DTU Orbit (02/12/2018)

The role of CO* as a spectator in CO2 electro-reduction on RuO2

RuO2-based electrocatalysts are found to be active at low overpotential toward direct electrochemical reduction of CO2 to formic acid and methanol. RuO2 can circumvent the thermodynamic bottleneck resulting from the scaling relations observed on metallic electrocatalysts, by utilizing an alternate pathway through oxygen-coordinated intermediates. Employing density functional theory based computational electrocatalysis models we show adsorbate–adsorbate interaction effects for adsorbates and reaction intermediates on the RuO2(110) surface are large and impactful to the reaction thermodynamics. We studied binding energy amendment due to adsorbate interaction (steric and electronic) with varying coverage of CO* spectators on the catalyst surface. Implications on the reaction pathways help us rationalize differences in experimentally observed carbonaceous product mix and suppression of the hydrogen evolution reaction (HER). We show that a moderate CO* coverage (~50%) is necessary for obtaining methanol as a product and that higher CO* coverages leads to very low overpotential for formic acid evolution. Our analysis also clarifies the importance of the reaction condition for CO2 reduction to liquid fuels utilizing RuO2-based electrocatalysts.

General information
State: Published
Organisations: Department of Energy Conversion and Storage, Atomic scale modelling and materials
Contributors: Bhowmik, A., Hansen, H. A., Vegge, T.
Pages: 18333-18343
Publication date: 2017
Peer-reviewed: Yes

Publication information
Journal: The Journal of Physical Chemistry Part C
Volume: 121
Issue number: 34
ISSN (Print): 1932-7447
Ratings:
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 4.58 SJR 2.135 SNIP 1.147
Web of Science (2017): Impact factor 4.484
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 4.48 SJR 1.964 SNIP 1.195
Web of Science (2016): Impact factor 4.536
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 4.68 SJR 1.886 SNIP 1.26
Web of Science (2015): Impact factor 4.509
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 5.08 SJR 2.032 SNIP 1.447
Web of Science (2014): Impact factor 4.772
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): CiteScore 5.14 SJR 2.143 SNIP 1.445
Web of Science (2013): Impact factor 4.835
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 4.98 SJR 2.529 SNIP 1.461
Web of Science (2012): Impact factor 4.814
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): CiteScore 4.92 SJR 2.339 SNIP 1.465