Simulation of Nanosystems for Energy Conversion

Master Thesis:

Carbon Neutral Fuels: CO2 Electro-Reduction to CH4 Simulated by a Kinetic Monte Carlo Model

Electrochemical CO2 reduction to CH4 (natural gas), which can be purely driven by renewable energies, is a highly promising catalytic reaction to synthesize carbon neutral fuels. However, economic commercialization of electrochemically synthesized CH4 is impeded by large required amount of electrical energy (arising from high overpotentials), energy dissipation toward the competing hydrogen evolution reaction (HER), and limited potential-dependent selectivity toward CH4 (see Figure 1).

With a focus on the catalytic processes under reactor environments at the nanoscale, kinetic Monte Carlo (kMC) simulations can contribute to a deeper understanding on how to reduce the overpotential and how to increase the selectivity toward CH4 at specific potentials, while suppressing the HER. In particular, beyond reproducing experimental results, this Master thesis can help to determine promising catalyst morphologies associated with suitable reactor environments such as temperature, gas pressures and bias potential.

Table 1. Faradaic Yields in CO_2 Reduction on Face-Centered Cubic (fcc) Metal Electrodes, As Reported by Hori et al.,[10] for Experiments at 5 mA cm^{-2} Current Density in a 0.1 M $KHCO_3$ Buffer at 18.5°C

		Faradaic yield, %				
electrode	V vs RHE	hydrocarbons/ organics[a]	CO	HCOOH	H_2	total
Ni	−1.09	2.1	0.0	1.4	88.9	92.4
Cu	−1.05	72.3	1.3	9.4	20.5	103.5
Pd	−0.81	2.9	28.3	2.8	26.2	60.2
Ag	−0.98	0.0	81.5	0.8	12.4	94.6
Pt	−0.68	0.0	0.0	0.1	95.7	95.8
Au	−0.75	0.0	87.1	0.7	10.2	98.0

[a]Predominantly CH_4, but also including C_2H_4, C_2H_5OH, C_3H_7OH, C_2H_6, C_3H_5OH, CH_3CHO, and C_2H_5CHO.

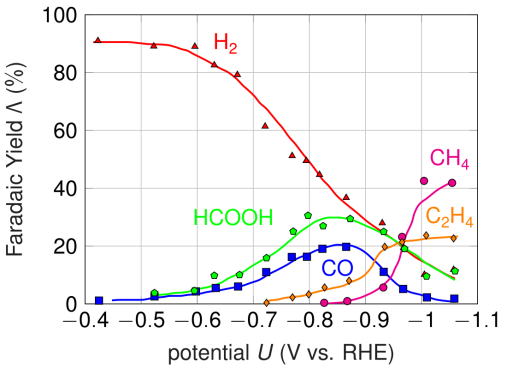

Figure 1: Selectivities from CO2 Reduction.
(Peterson, Noskov, J. Phys. Chem. Lett., 3, 2012; Hori et al., J. Chem. Soc., 8, 1989, *adapted*)