UniSysCat Colloquium

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Start Time: Wednesday, September 16, 2020 05:00 pm

End Time: Wednesday, September 16, 2020 06:00 pm

Zoom Meeting
Technische Universität Berlin, Straße des 17. Juni 115, 10623 Berlin

The search for a new Deacon catalyst: modeling catalyst stability across the periodic table

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Catalysts degrade over time, which means that they lose catalytic activity or selectivity due to a variety of aging processes. While catalyst screening (both experimental and theoretical) regarding activity and selectivity is well-established in catalysis research, screening with respect to stability is experimentally difficult due to the long time scales of catalyst degradation. Computational catalyst screening can complement experimental studies aiming at the identification of new catalyst materials that are stable under operating conditions.

In the HCl oxidation in the Deacon process using an oxide catalyst MO_x, the main catalyst degradation mechanisms are catalyst leaching in the form of volatile M-O-Cl compounds, as well as bulk phase transformation of oxide catalysts into less catalytically active chlorides. We present a catalyst screening approach based on experimental thermodynamic data for the compounds in the M-O-Cl system. In this approach, we divide the M-O-Cl system into four groups of compounds, oxides, chlorides, condensed and gaseous species.

It can address specific reaction conditions by computing the driving forces (ΔG) of chlorination and sublimation as a function of temperature, reactant feed composition and conversion. We apply it to the M-O-Cl compounds of 66 elements, regarding chlorination and volatility as separate descriptors. According to previous experimental studies, catalyst degradation starts at the reactor inlet and is driven by water formation. At the target reaction conditions (T < 650 K,

dry, $p(HCl)/p(O_2) \approx 1$, conversion < 1 %), 13 oxides are identified as suitable candidates. While some of the candidates (TiO_2, ZrO_2, Al_2O_3, and SiO_2) are inactive (but already employed as catalyst supports in the Deacon process), other candidates are ruled out for safety concerns (BeO, ThO_2, UO_3). Catalytic activity of the remaining candidates is assessed preliminarily by employing the oxygen and binding energies at the surface as a descriptor.

Dr. Ingo Zebger

Organizer