Decoupling of redox/basic catalytic mechanisms during n-Butanol production
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Key results—catalytic performance
- Effects brought by the introduction of redox sites
- Relationship between active sites and product selectivity

Observations:
- Redox sites greatly increased reaction rate and suppressed acid-catalyzed products.
- Redox sites negatively affect BuOH selectivity. MoO3 showed the best potential as promoter.

Proposed active site(s) configuration in MoO3/MgAlO catalyst

Levels of acidity

Lewis acidic sites

Proposed active sites: $\text{MoO}_3$ $\text{MgAlO}$ catalyst

Key results—Qualification and quantification of catalytic relevant basic sites
Basic sites: necessary for enolate formation from acetaldehyde intermediates (generated from EtOH) to realize C-C coupling (key step for BuOH formation).

Method I: Acetic acid competitive adsorption—Reactive basic sites

Method II: TGA of CO adsorption/desorption—Total basic sites

Method III: Deconvolution of CO$_2$-TGA results—Types and amount of different basic sites

Key results—In situ IR spectra

- Identifying basic sites with different strengths

CO$_2$ desorption from 298 to 800 °C: Monitor the IR band of adsorbed CO$_2$ surface species on various basic sites (different carbonate forms)$^{[2]}$

- Identifying the basic sites for C-C coupling step

Key conclusions
- Introduced redox functionalities can increase reaction rates by activating the hydrogen-transfer steps.
- n-Butanol formation is limited by the C-C coupling step.
- Strong basic sites are the active center to realize the C-C coupling.

Reference

Acknowledgements

Motivation and background
- n-Butanol is widely used as an important industrial intermediate. Based on the combustion performance it is a superior alternative biofuel compared to bioethanol$^{[1]}$.
- The large supply of bioethanol on the market, justifies seeking a green and efficient route for the direct catalytic conversion of ethanol (EtOH) to n-butanol (BuOH).
- Current technology for this route is still limited to bench scale due to the lack of suitable catalysts and optimized reaction systems$^{[2]}$.

Objectives
- Regulating the catalytic activity of MgAlO-based catalysts to optimize n-butanol production.
- Develop methodology to identify and qualitatively and quantitatively correlate the relationships between active sites and catalytic performance.

Research Methodology and equipment
- Reaction network of the EtOH to BuOH process
- Design of MgAlO-based functional catalysts
- Operando spectroscopy characterizations system

Chemical and Biochemical Engineering

Butanol formation is limited by the C-C coupling step. In situ IR spectra show increased reaction rates by activating hydrogen-transfer steps.