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SELECTIVE ETHANOL OXIDATION ON SUPPORTED BIMETALLIC GOLD CATALYSTS: BASE CHEMICALS FROM "GREEN" PROCESSES

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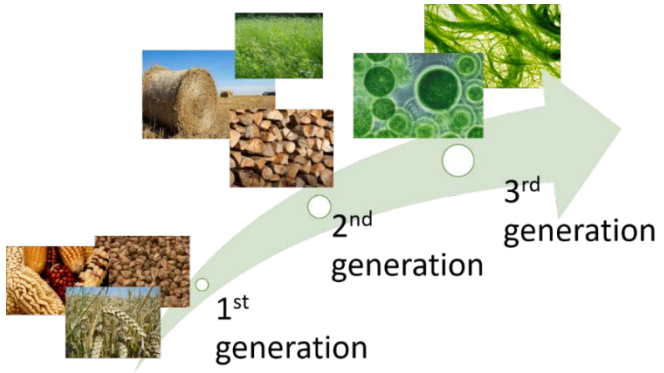
7/8 June 2018

Motivation

Bioethanol as Feedstock

- Green Chemistry: Biomass as sustainable substitute for oil-based economy
- Selective oxidation of Ethanol: **Acetaldehyde** and Acetic Acid

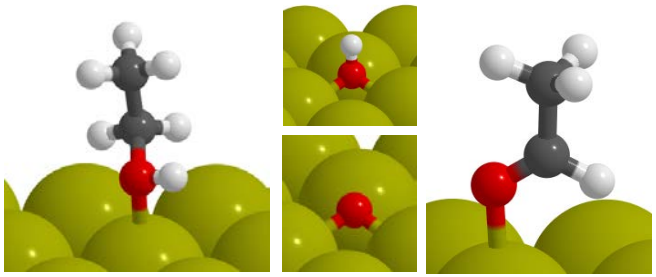


Au catalysts with promoters (Ag, Ru, etc.) for enhancing oxygen activation.

Fundamental Interest in Bimetallic Catalysts

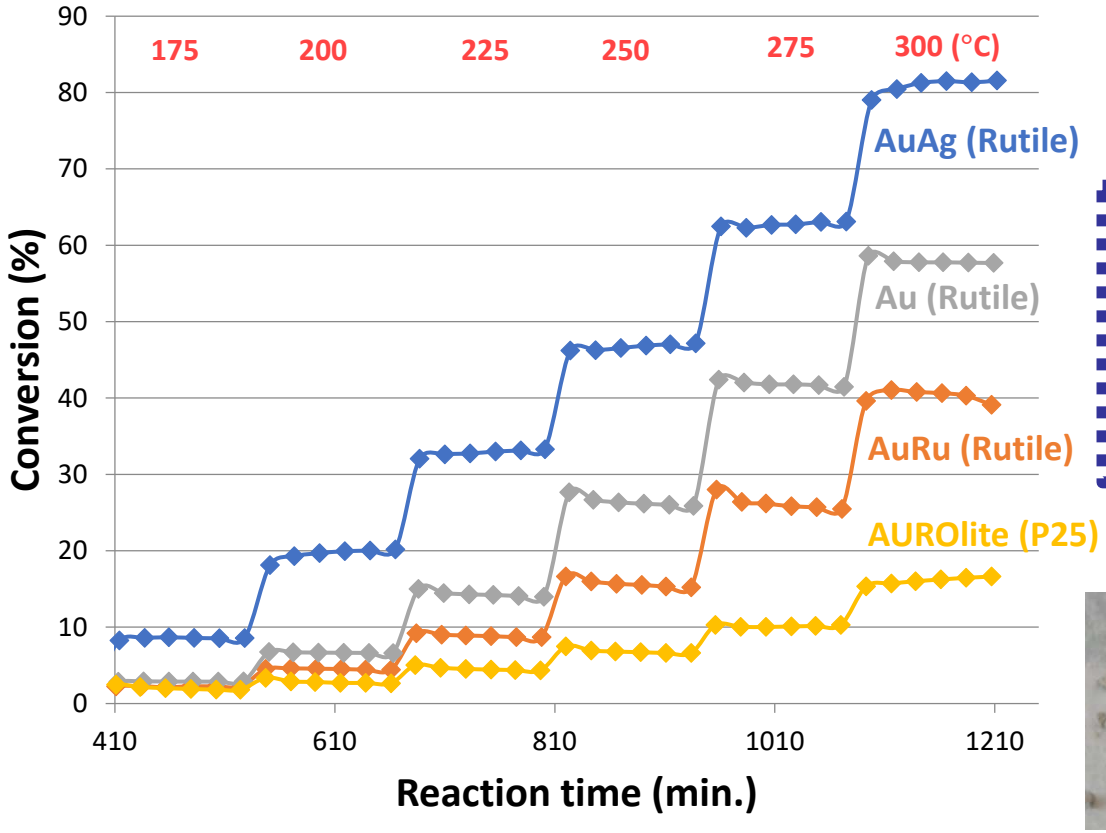
- Potentially tunable properties
- Choice of second metal component
- Synergistic effect of two metal components

EtOH (O/OH) → Alkoxy (O/OH)→ Acetaldehyde



Theoretical DFT calculations by Prof. Saeys and Jenoff de Vrieze, University of Gent.

Catalytic Properties



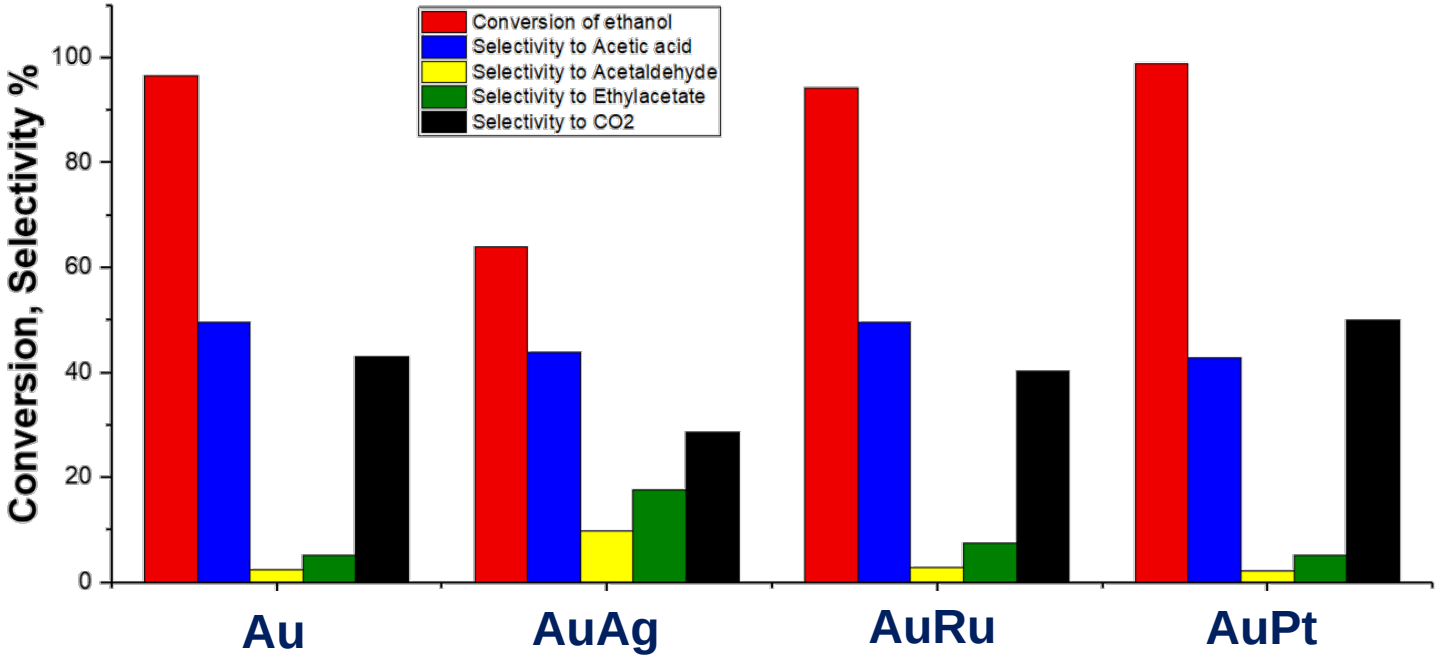
Gas-phase reaction!

Selectivity: > 95%!

- **Best catalysts:**
  AuAg > Au > AuRu > AUROlite (commercial reference)
- **Support:**
  TiO_2: Rutile (AUROlite: P25 = mixture of Rutile and Anatase)

Catalytic Properties

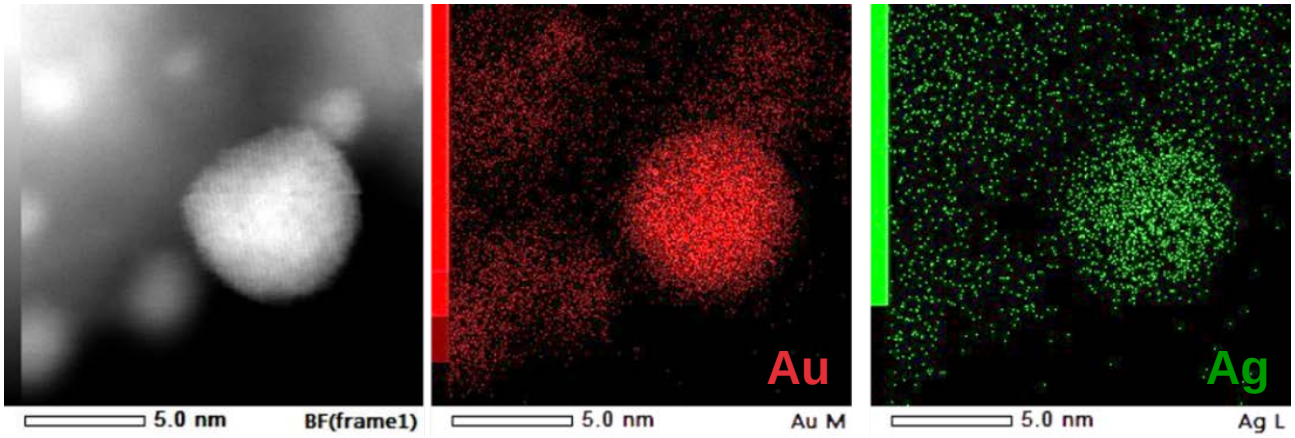
Liquid phase reaction!



- **Activity trend reversed:**
  Gas: AuAg > Au > AuRu > AuPt
  Liquid: AuAg < AuRu < Au < AuPt

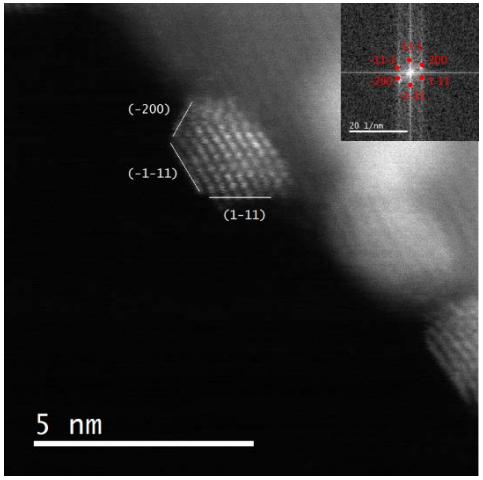
In cooperation with F. Robles-Méndez, S. Mostrou-Moser, J. v. Bokhoven, ETH Zürich.

Catalyst Material: AuAg

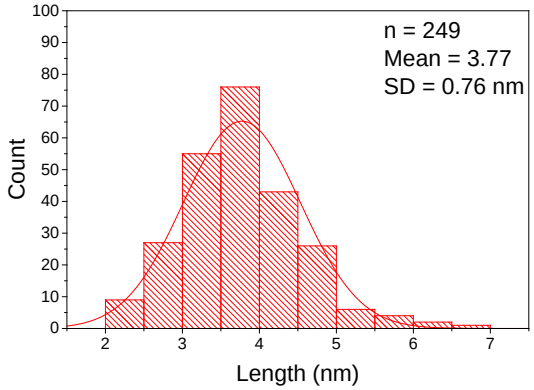


In cooperation with G. Dražić, National Institute of Chemistry, Ljubljana.

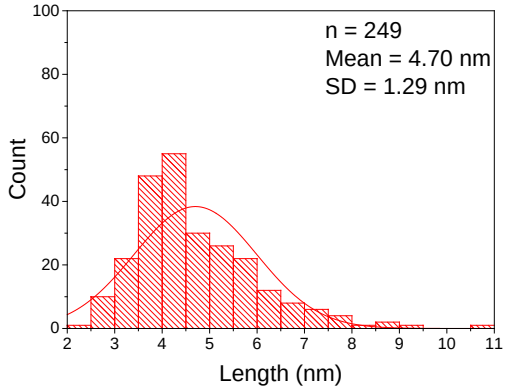
(Cs-corrected) HR-STEM/STEM-EDX: Field Emission Scanning Electron Microscope (FE-SEM) model JSM-7000F



After pretreatment

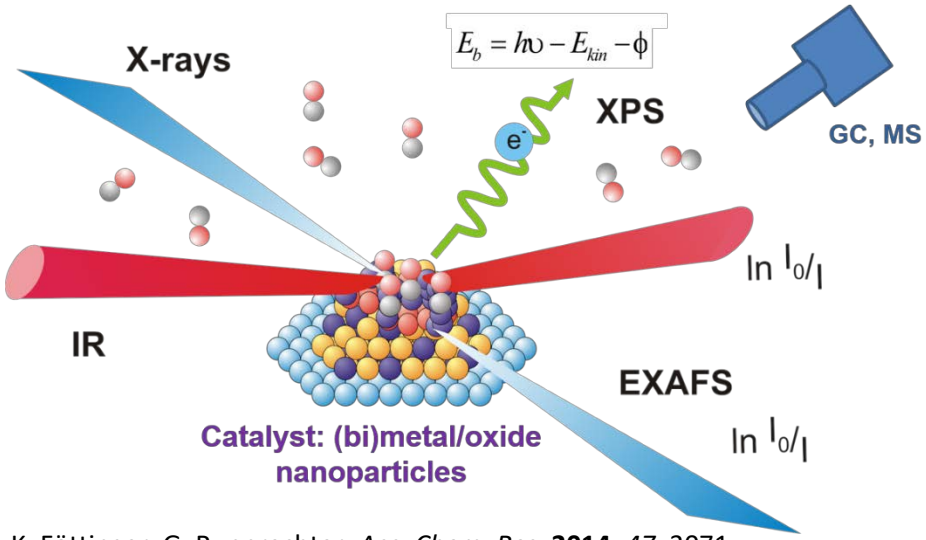


After reaction



Measured at FEI Tecnai F20 (STEM-HAADF) at 200 kV at USTEM, TU Wien.

Operando Spectroscopy



K. Föttinger, G. Rupprechter, *Acc. Chem. Res.* **2014**, *47*, 3071.

Near Ambient Pressure (NAPP)-XPS

- **Bessy, ISISS Berlin:** 30 April – 7 May
- **ALBA, CIRCE Barcelona:** 29 May – 4 June

EXAFS

- **DESY, P64 Hamburg:**17 May – 19 May
- **ALBA, CLÆSS Barcelona**: 22 May – 28 May
- **DLS, B18 (Didcot/UK):** 11 July – 14 July

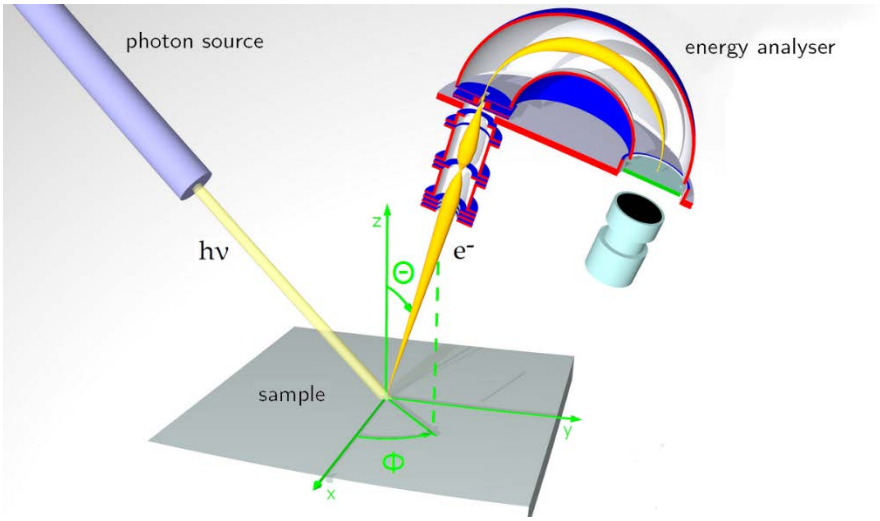
DRIFTS/IR:

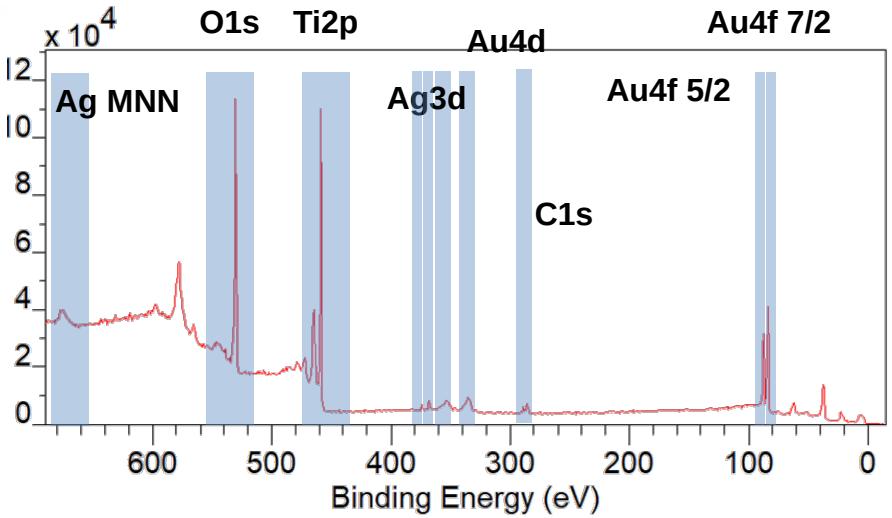
TU Wien

Raman:

Groningen/NL

Operando XPS

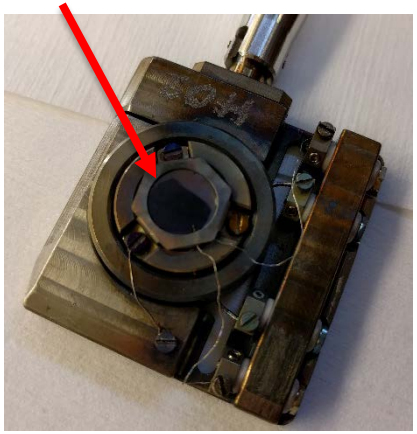


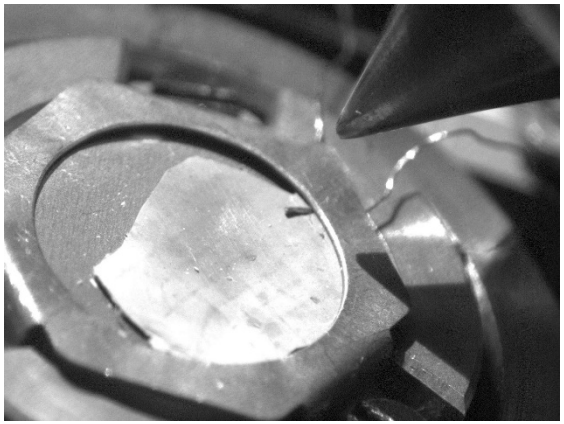


Surface-sensitive element information!

Sample

Sample

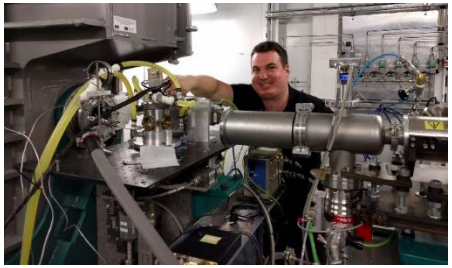


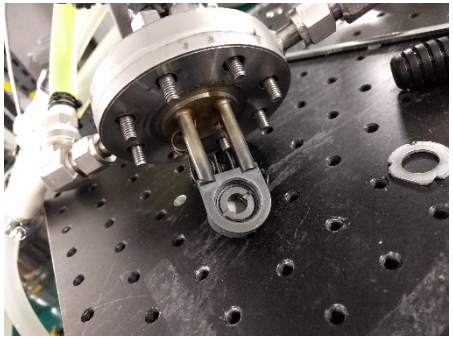


Oxidation state

Depth profiling

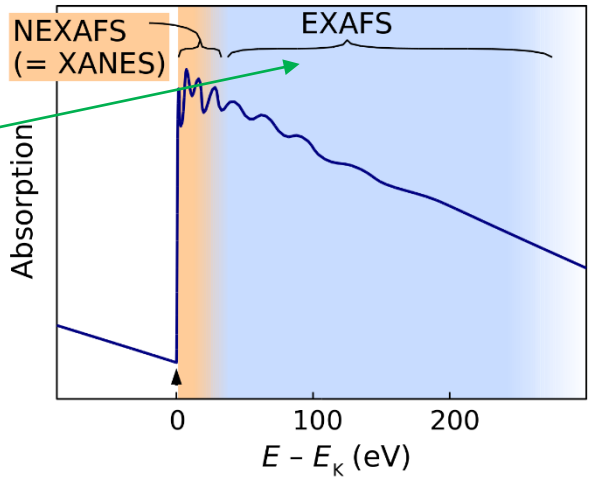
Operando EXAFS



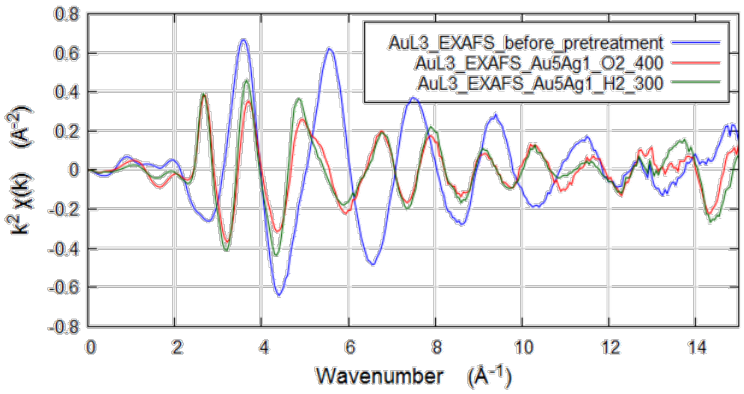


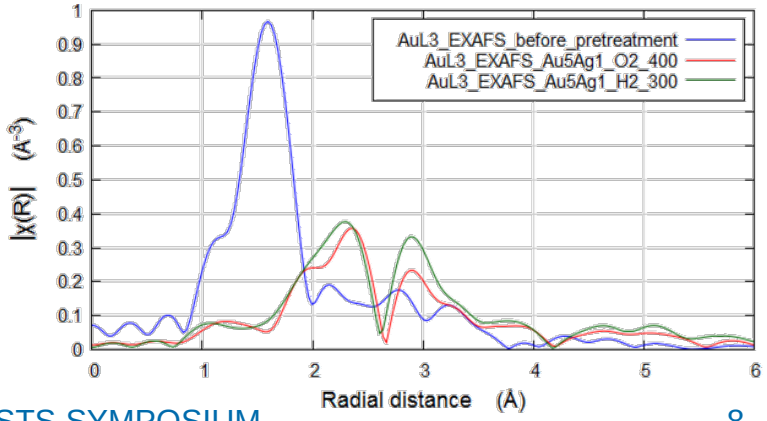
Chemical Information →

Geometry, Structure →



- Formation of nanoparticles.
- First evidence of alloying!
- (Partial!) oxidation of Ag.





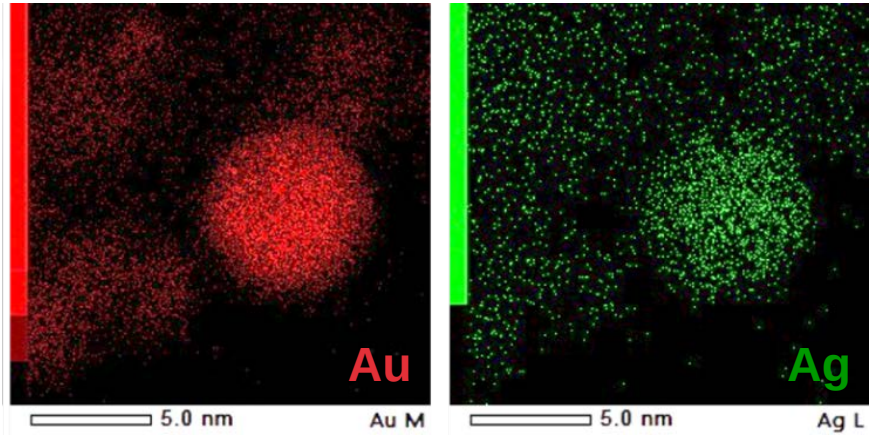
Conclusion and Outlook

Kinetics

- AuAg is a promising catalyst for selective EtOH oxidation.
- Selectivity: > 95%, Side products: Ethyl acetate, Acetic acid, (CO_2)
- Synergistic effect between Au and Ag!
- Opposite trend gas-liquid:
  Gas: AuAg > Au > AuRu > AuPt
  Aqueous: AuAg < AuRu < Au < AuPt

Characterization

- Bimetallic particles – alloying!
- (Partial) oxidation of Ag.
- First evidence of Ag segregation on the surface.



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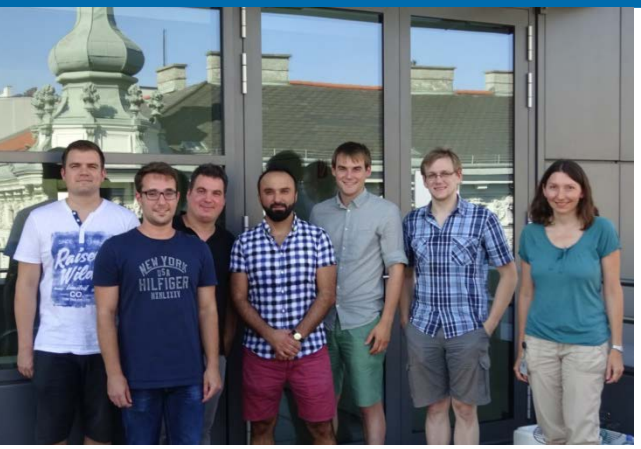
Vortrag NSM.24



FWF Der Wissenschaftsfonds.

LIONS INTERNATIONAL

ETH zürich

HZB Helmholtz Zentrum Berlin

ALBA

DESY.

UNIVERSITEIT GENT

USTEM

diamond

TU WIEN aic analytical instrumentation center

NATIONAL INSTITUTE OF CHEMISTRY

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