Molybdenum sulfide-derived molecular cluster complexes for direct preparation of benzimidazoles

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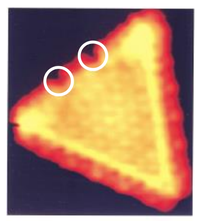
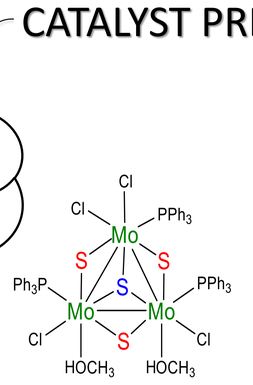
MOLYBDENUM DISULFIDE



GOAL
Presence of active sites in edges and basal planes

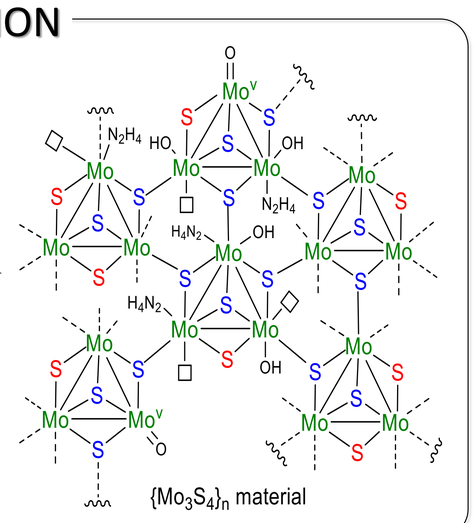
CATALYTIC ACTIVITY
- ❑ Structural defects: S vacancies
- ❑ Metallic electronic states
- ❑ Inactive basal planes

CATALYST PREPARATION



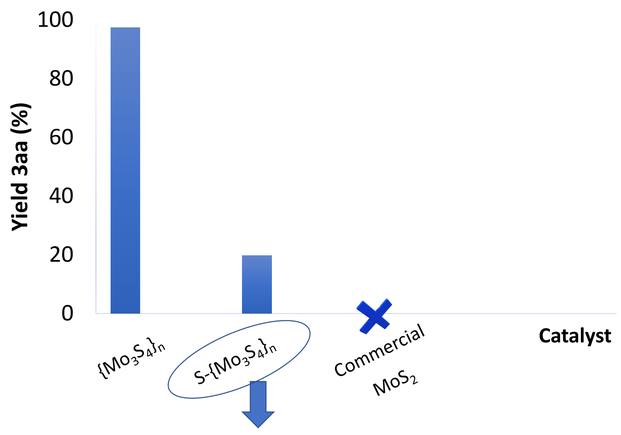
Mo_3S_4 molecular complex

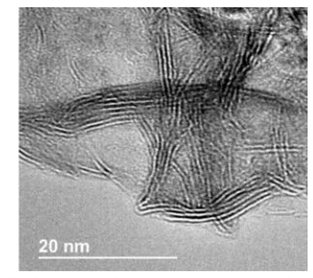
$H_2N\text{-}NH_2$, H_2O



$\{Mo_3S_4\}_n$ material

CATALYTIC ACTIVITY



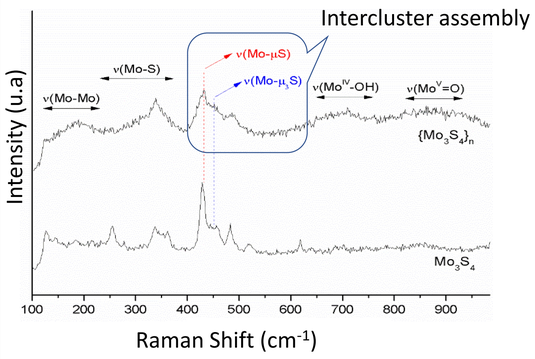


Morphological characterization of S-$\{Mo_3S_4\}_n$

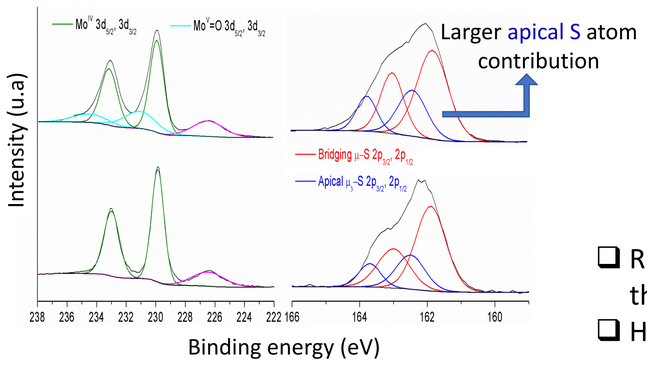
- ❑ Stacked layered structure
- ❑ Low number of active sites in their basal planes
- ❑ Poor catalytic activity

CATALYST CHARACTERIZATION

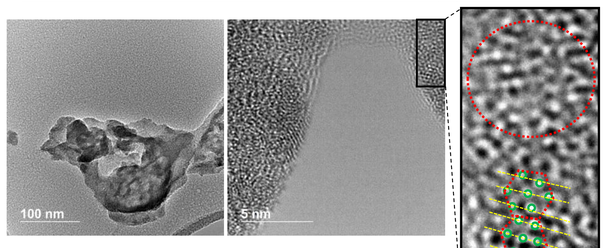
Raman Spectroscopy



X-ray Photoelectron Spectroscopy (XPS)



Transmission Electron Microscopy (TEM)

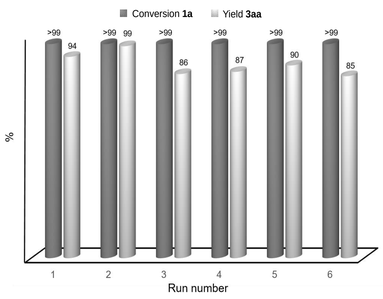


- ❑ Randomly agglomerated nanosheets that preferentially expose their basal planes
- ❑ High structural disorder

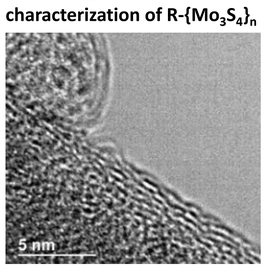
↳ **GOOD CATALYTIC ACTIVITY**

SCOPE

CATALYST RECYCLING

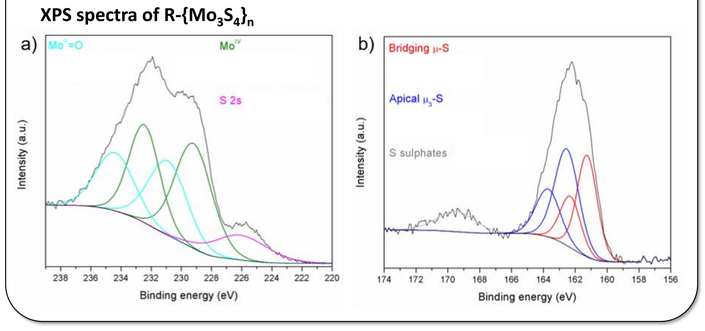


Morphological characterization of R-$\{Mo_3S_4\}_n$

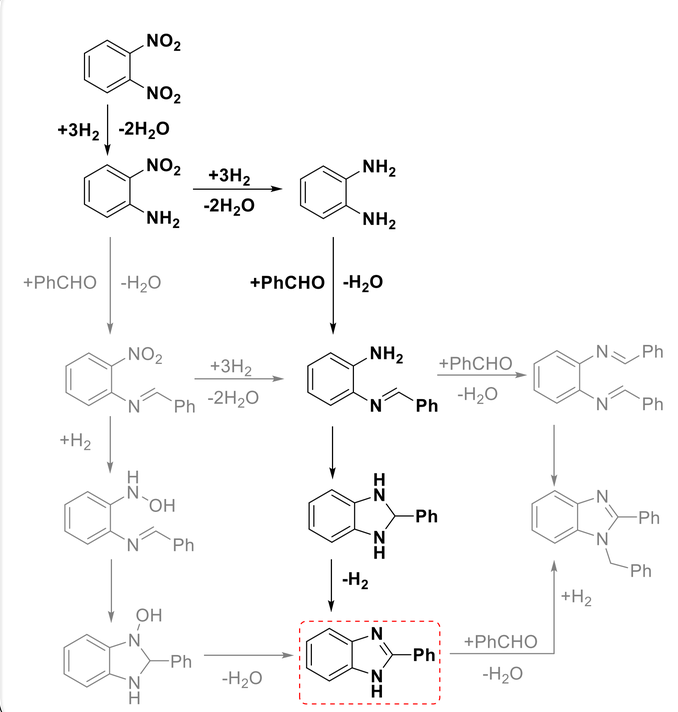


PRESERVATION OF THE MOLECULAR CLUSTER STRUCTURE

XPS spectra of R-$\{Mo_3S_4\}_n$



PROPOSED MECHANISM



CONCLUSIONS

Innovative Catalyst Synthetic Strategy:
- ❑ Based on molecular cluster complexes
- ❑ Creation of structural defects in basal planes
- ❑ Enhanced catalytic activity
- ❑ Good recycling

Simple and Direct Synthesis of Benzimidazoles:
- ❑ Broad scope and high selectivity
- ❑ Use of a green reductant: H_2
- ❑ High atomic efficiency
- ❑ Non-noble metal-based catalyst

REFERENCE: Miriam Rodenes, Francisco Gonell, Santiago Martín, Avelino Corma, Iván Sorribes. Molecularly Engineering Defective Basal Planes in Molybdenum Sulfide for the Direct Synthesis of Benzimidazoles by Reductive Coupling of Dinitroarenes with Aldehydes. *JACS Au* **2022**, 2, 601-612.