

Investigation on the Formation of Negatively Charged Gold Species on SiO₂ and the Activity in CO Oxidation

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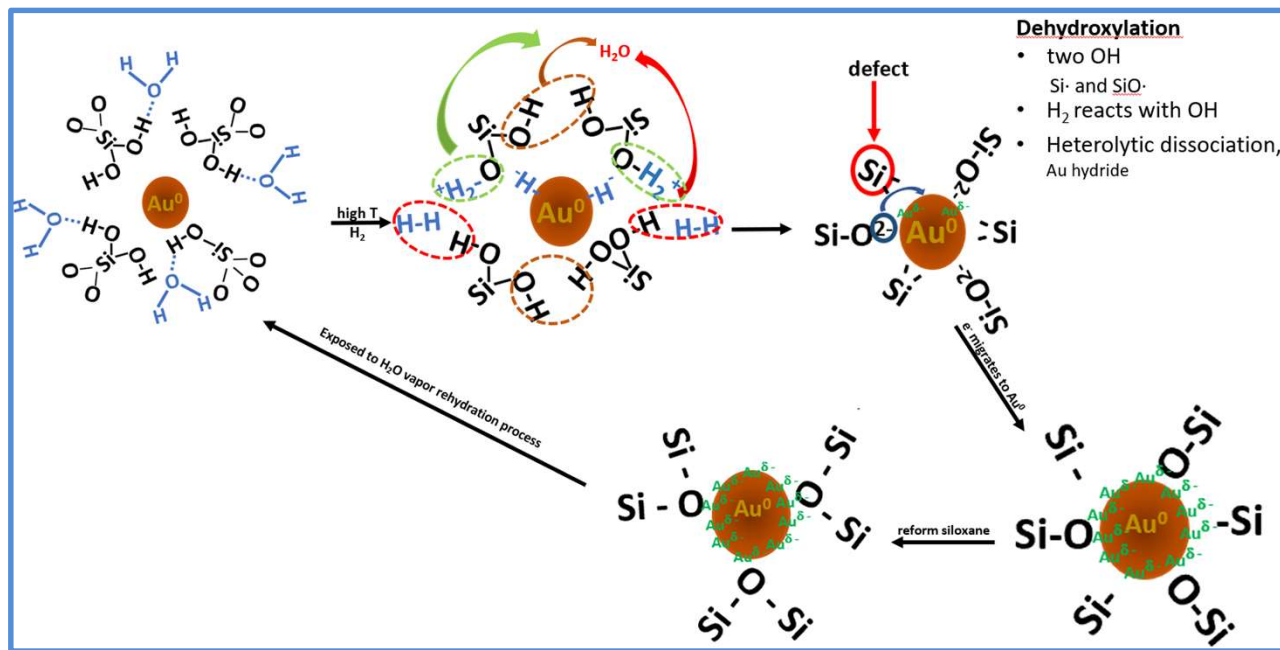
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Project Overview

- CO-FTIR: Au^{δ+} (> 2120cm⁻¹), Au⁰ (2120-2080cm⁻¹), Au^{δ-} (<2080cm⁻¹);
- Au^{δ-} species observed on **nonreducible supports** (SiO₂, Al₂O₃) with various thermochemical treatments (H₂ & N₂, 180⁰C, 240⁰C, 290⁰C, 400⁰C);
- Activity of Au^{δ-} species on room temperature **CO oxidation**.

Hypothesis



e⁻ on Au^{δ-} from:

- Active defects
≡Si·, ≡Si-O·
- Au hydride

Methods

Materials: Aerosil 300 (SA ~300m²/g), gold ethylenediamine;

Synthesis: SEA & DI, 2.2wt%Au, reduced @ 400⁰C.

Particle size: SEA-1.6nm, DI-11.2nm;

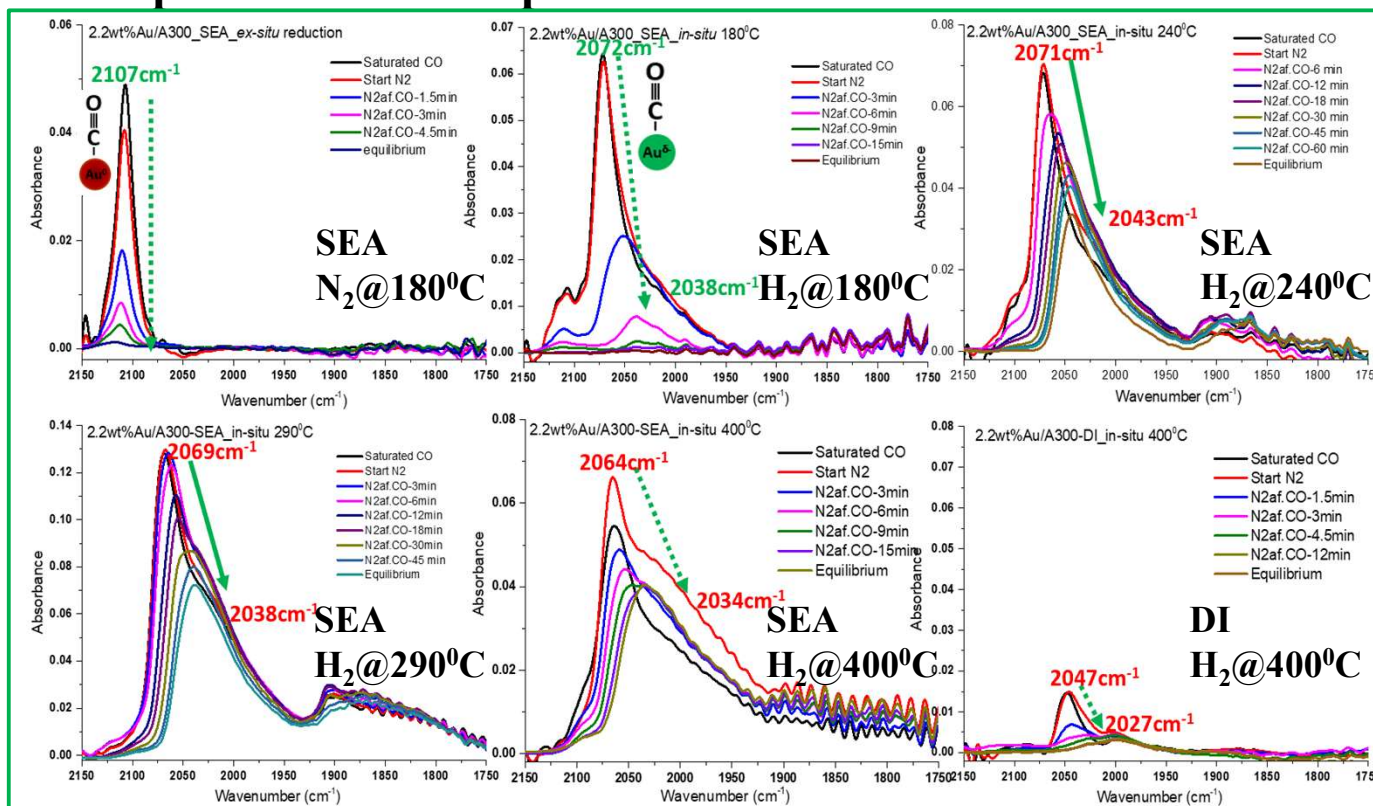
Characterization: CO-FTIR to check the valent states of Au;

Evaluation: Vertical gas flow reactor for RT CO oxidation.



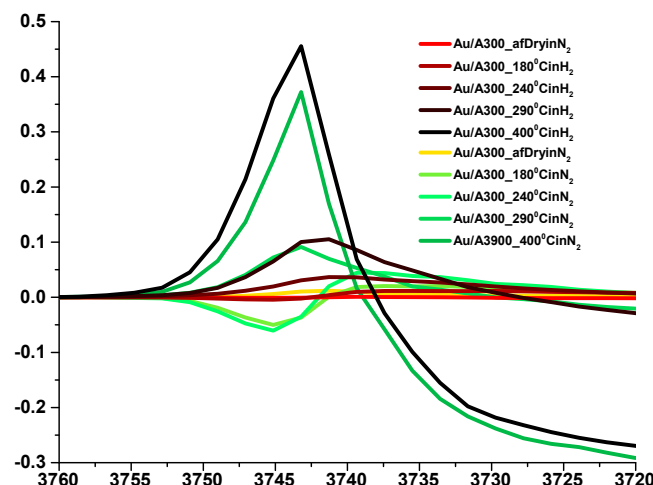
Results and Discussion

---IR spectra for Au samples with various thermochemical treatments



Au⁰ (2120-2080cm⁻¹);
Au^{δ-}(<2080cm⁻¹)

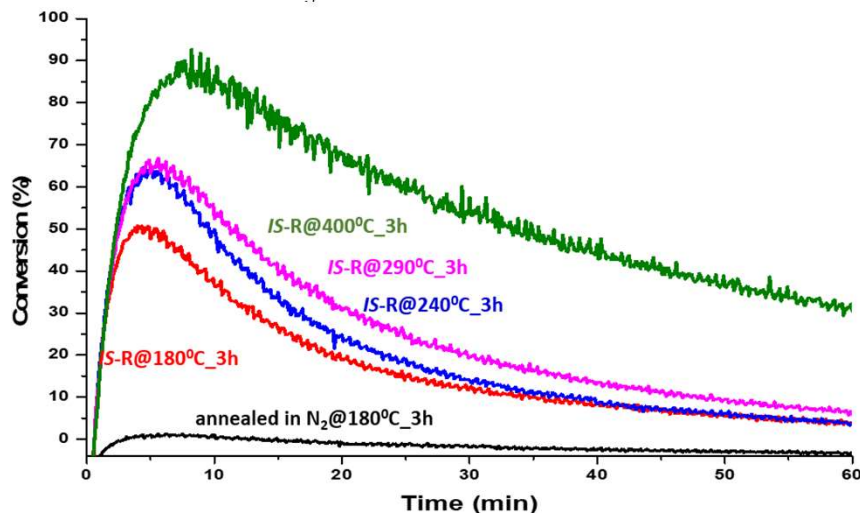
---IR spectra for OH region



- H₂ enhanced de-hydroxylation of SiO₂.

- H₂, high T, small NPs

--- Activity in CO Oxidation



- Higher in-situ reduction T led to a higher initial activity;
- Rapid deactivation is caused by the consumption of Au^{δ-} species or carbon accumulation.



Conclusions

- Small NPs, high T and H_2 is necessary for the formation of $Au^{\delta-}$ on SiO_2 ;
- Electrons on the active defective sites ($Si\cdot$, $SiO\cdot$) produced during the dehydroxylation of SiO_2 or on Au hydride can transfer to Au^0 and formed $Au^{\delta-}$ species;
- Increased activity in RT CO oxidation is achieved over Au sample in-situ reduced at a higher T; rapid deactivation is caused by the consumption of $Au^{\delta-}$ species.