UNIVERSITEIT GENT

FACULTY OF **ENGINEERING AND ARCHITECTURE**

# Total oxidation of toluene over $CuO\text{-}CeO_2/Al_2O_3$: reaction network and catalyst characterization


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M2dcR2

## Introduction

Catalytic total oxidation is the most promising approach for the abatement of Volatile Organic Compounds (VOCs) owing to its high efficiency and low operating temperature. Copper promoted by ceria shows high catalytic performance for the complete oxidation of VOCs (toluene, propane, benzene).

The central issues addressed in this study are: (1) reaction network of toluene total oxidation, (2) the nature of the active sites of the CuO – $CeO_2$ catalyst, (3) participation of $CeO_2$ in the oxidation of reduced copper, (4) the role of $CeO_2$ in the enhancement of catalytic activity in the presence of water and carbon dioxide.

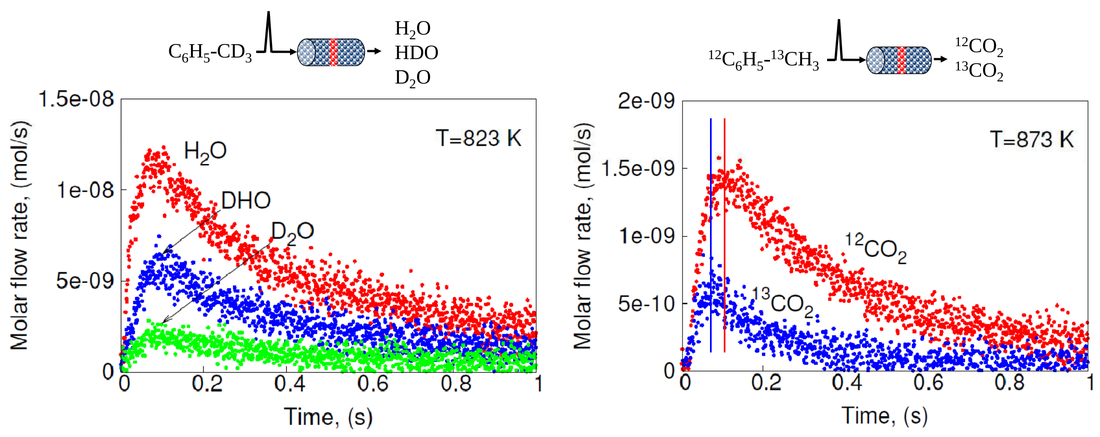
## Experimental

TAP

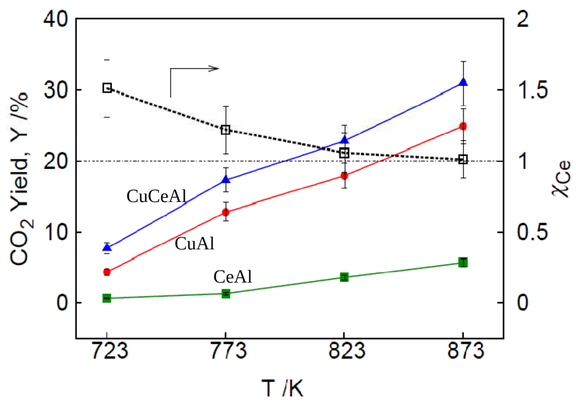

TEM

EXAFS

## Isotopic labeling experiments



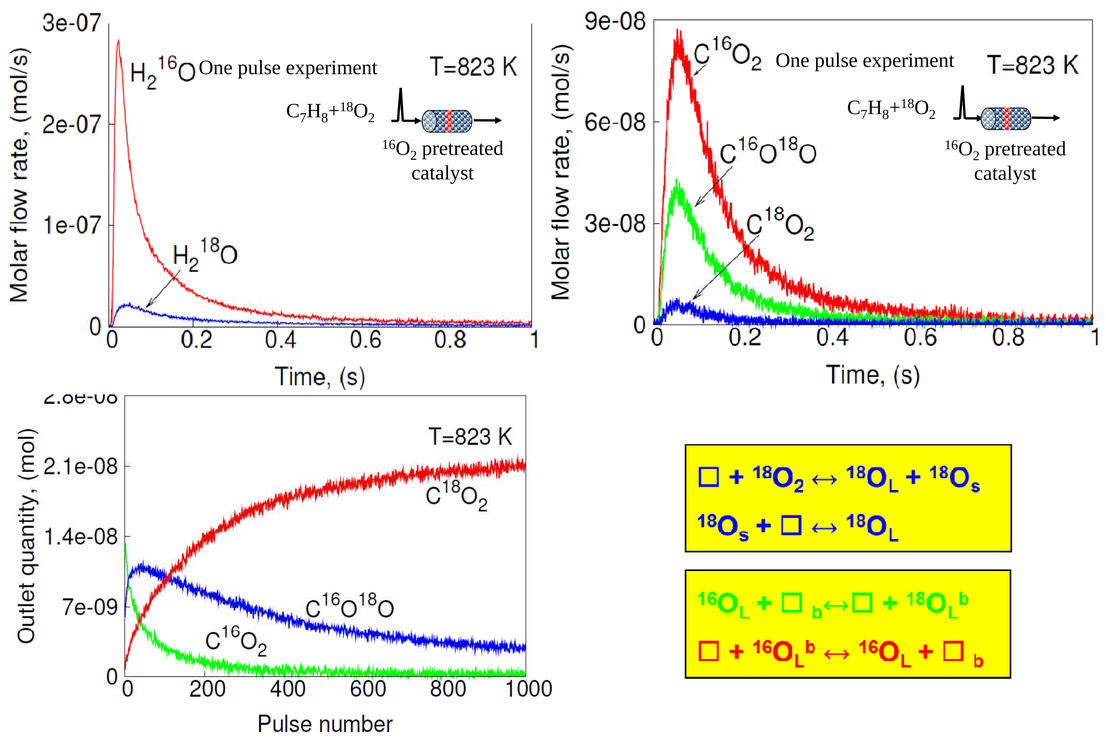

## Catalytic performance of single and binary metal oxides




The promoting or inhibiting effect of $CeO_2$ in $CuO\text{-}CeO_2/Al_2O_3$

$$\chi_{Ce} = \frac{Y_{CuO-CeO_2/Al_2O_3}}{\left(Y_{CuO/Al_2O_3} + Y_{CeO_2/Al_2O_3}\right)}$$

## Oxygen isotopic exchange experiments



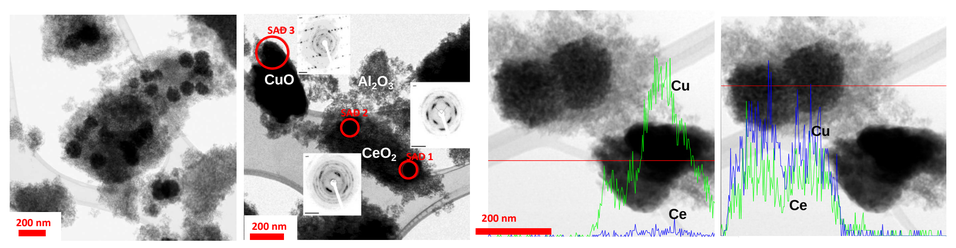

□ + $^{18}O_2$ ↔ $^{18}O_L$ + $^{18}O_s$

$^{18}O_s$ + □ ↔ $^{18}O_L$

$^{16}O_L$ + □ $_b$↔□ + $^{18}O_L{}^b$

□ + $^{16}O_L{}^b$ ↔ $^{16}O_L$ + □ $_b$

## Microstructure characterization

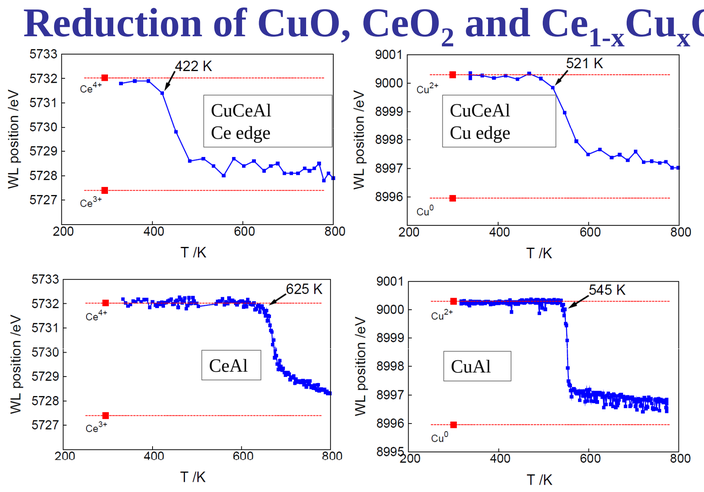



HRTEM

SAED

EDX

## Reduction of CuO, $CeO_2$ and $Ce_{1-x}Cu_xO_2$

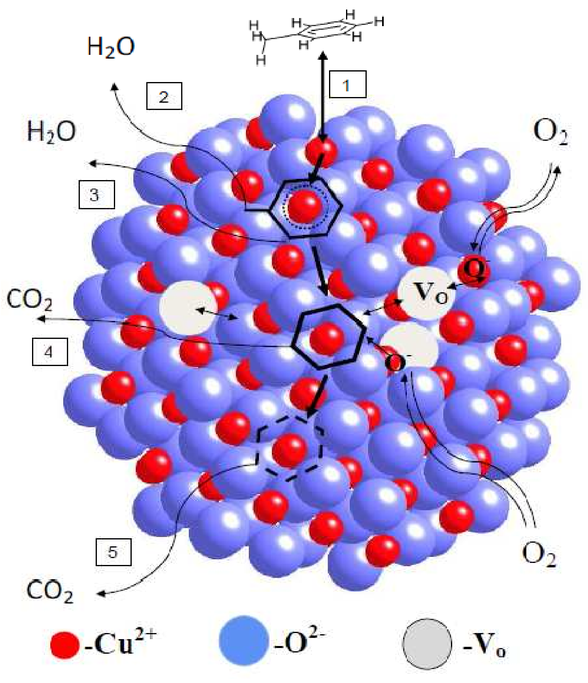



White line energy position vs. temperature during hydrogen reduction of $CuO\text{-}CeO_2/Al_2O_3$; $CeO_2/Al_2O_3$; $CuO/Al_2O_3$

Ce $L_{III}$ edge; Cu K edge.

$Ce^{4+}$ reduces first, passing O to nearby Cu, thus keeping it oxidized.

## Conclusions




### Reaction network

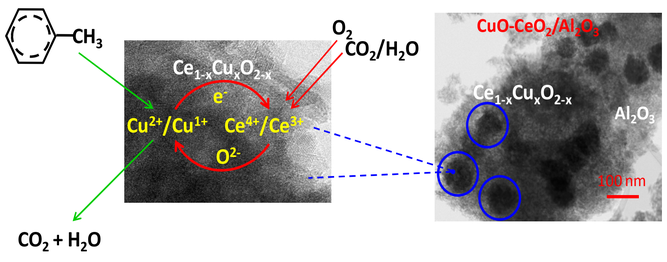
**Step 1.** adsorption of toluene

**Steps 2 and 3**. H-abstraction from the methyl groups and phenyl groups and water formation

**Steps 4 and 5**. abstraction of the carbon atom in the methyl group and phenyl group and formation of $CO_2$

## Conclusions

Enhanced activity is due to the solid solution

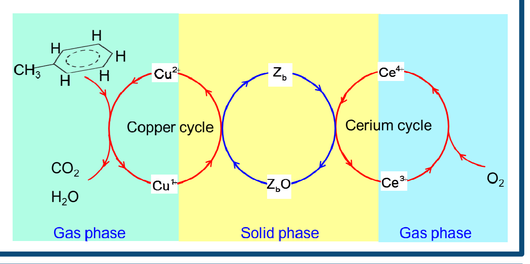



**Tol + $Cu^{2+}$ → $Cu^{1+}$ + $CO_2$ + $H_2O$**

**$Ce^{4+}$ + $Cu^{1+}$ → $Ce^{3+}$ + $Cu^{2+}$**

**$2Ce^{3+}$ + $O_2$ → $2Ce^{4+}$**

**$Ce^{3+}$ + $H_2O/CO_2$ → $Ce^{4+}$ + $H_2/CO$**





### References

[1] Unmesh Menon, Vladimir V. Galvita, and G.B. Marin, J. Catal. 283 (2011) 1-9

[2] U. Menon, V.V. Galvita, H. Poelman, V. Bliznuk, D. Poelman, G.B. Marin, Submitted to J. Catal. (2012)



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