6WCOC, Lille, France, July 5-10, 2009

UNIVERSITEIT GENT

FACULTY OF ENGINEERING

TAP-study on the total oxidation of propane over a $CuO\text{-}CeO_2/\gamma\text{-}Al_2O_3$ catalyst

V. Balcaen, R. Roelant, H. Poelman, D. Poelman, G.B. Marin

Laboratory for Chemical Technology

IDECAT Integrated Design of Catalytic Nanomaterials for a Sustainable Production

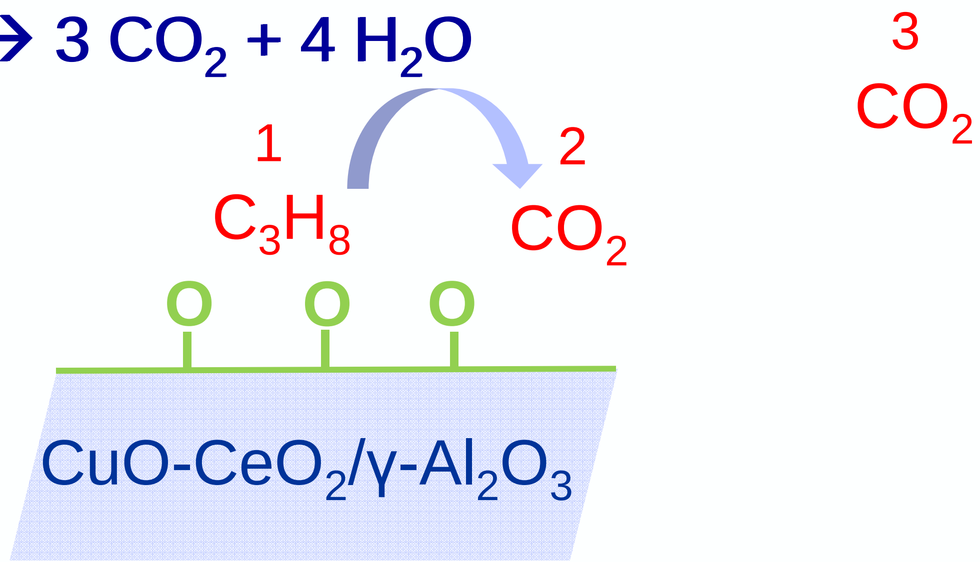
http://www.lct.ugent.be

Introduction

- VOCs = Volatile Organic Compounds → important air pollutants

→ Total catalytic oxidation

$$C_3H_8 + 5\ O_2 \rightarrow 3\ CO_2 + 4\ H_2O$$



1. Which species are responsible for converting propane to CO_2?
2. What is the role of the different metal oxide phases?

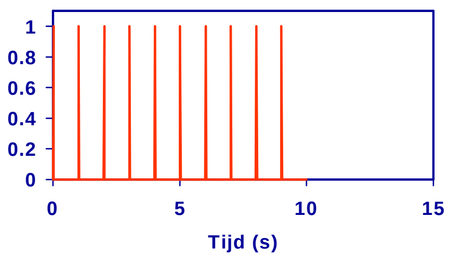
Outline

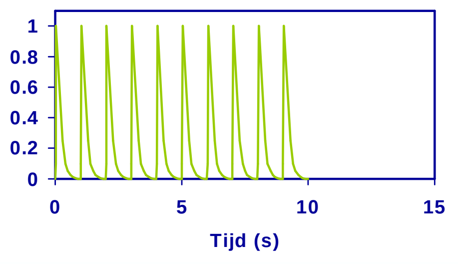
1. Introduction
2. Experimental set-up, conditions and catalysts
3. Results
    - Role and nature of active oxygen species
    - Oxygen mobility
    - Role of metal oxides
4. Conclusions

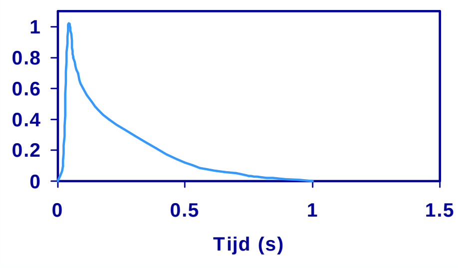
Three types of TAP pulse experiments

Inlet | **Outlet** | **Response**

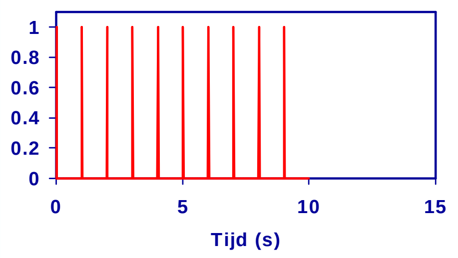
Single-pulse = State-defining

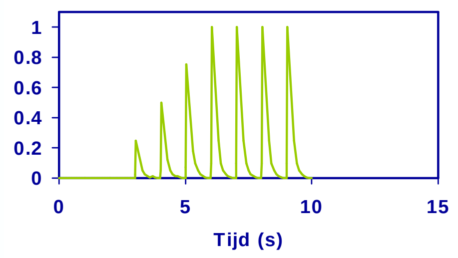


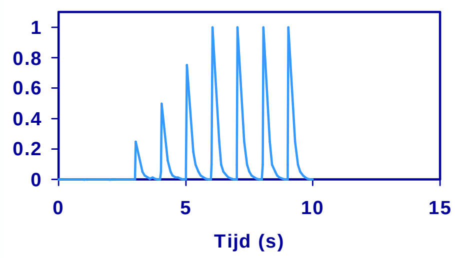




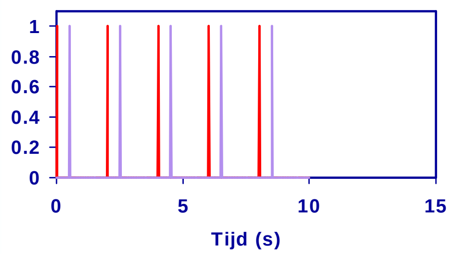
Multi-pulse = State-altering

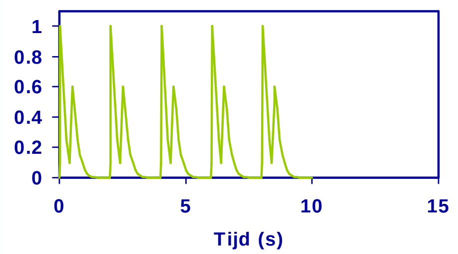


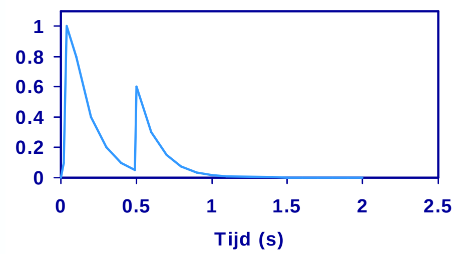




Alternating pulse

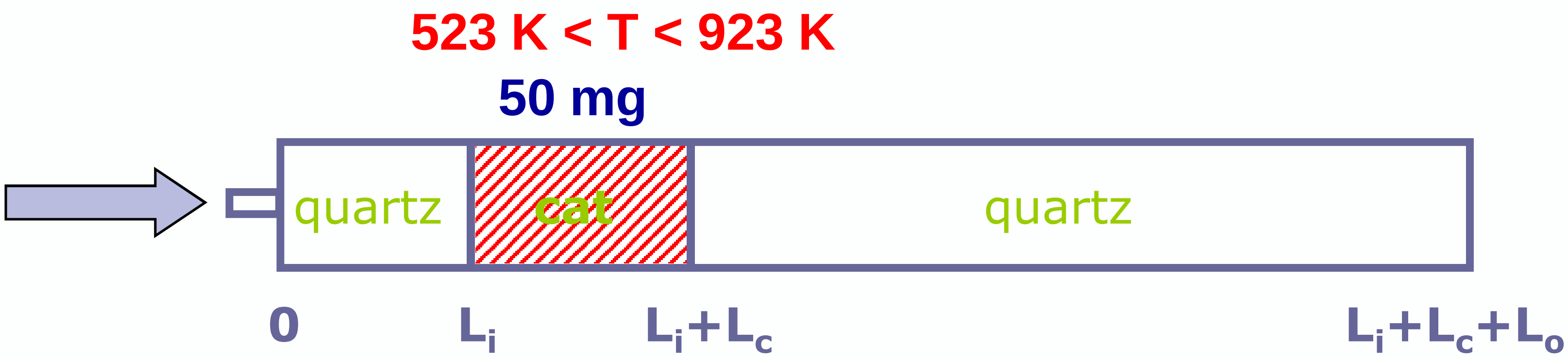






Experimental conditions

C_3H_8 / Kr
O_2 / Ar
CO_2 / Ar
$C^{18}O_2$ / Ar

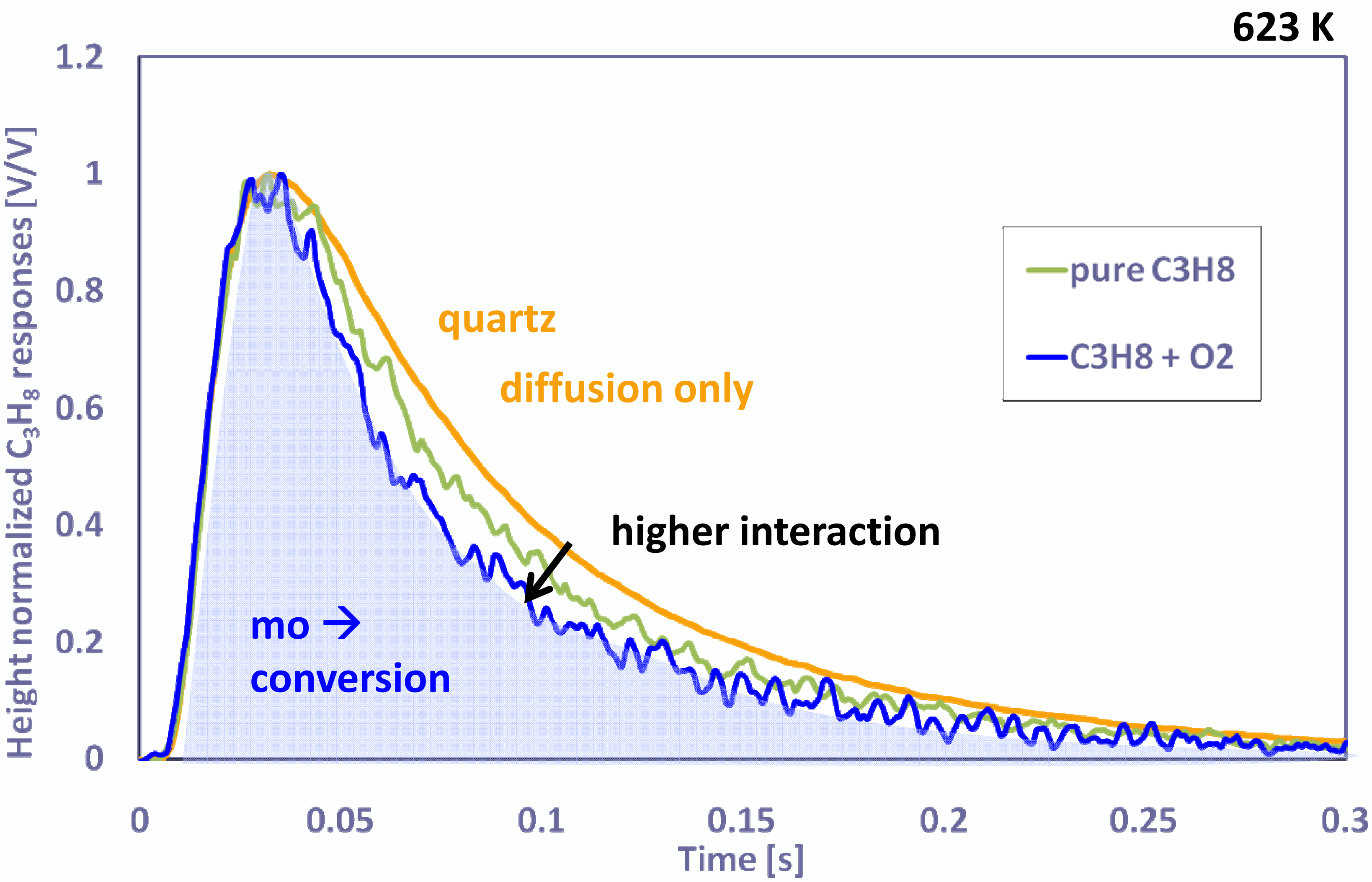


O atoms related to	CuO and/or CeO_2 (10^{19})	Al_2O_3 (10^{19})
CuO-CeO_2/γ-Al_2O_3	7	70
CeO_2/γ-Al_2O_3	2	80
CuO/θ-Al_2O_3	5	70

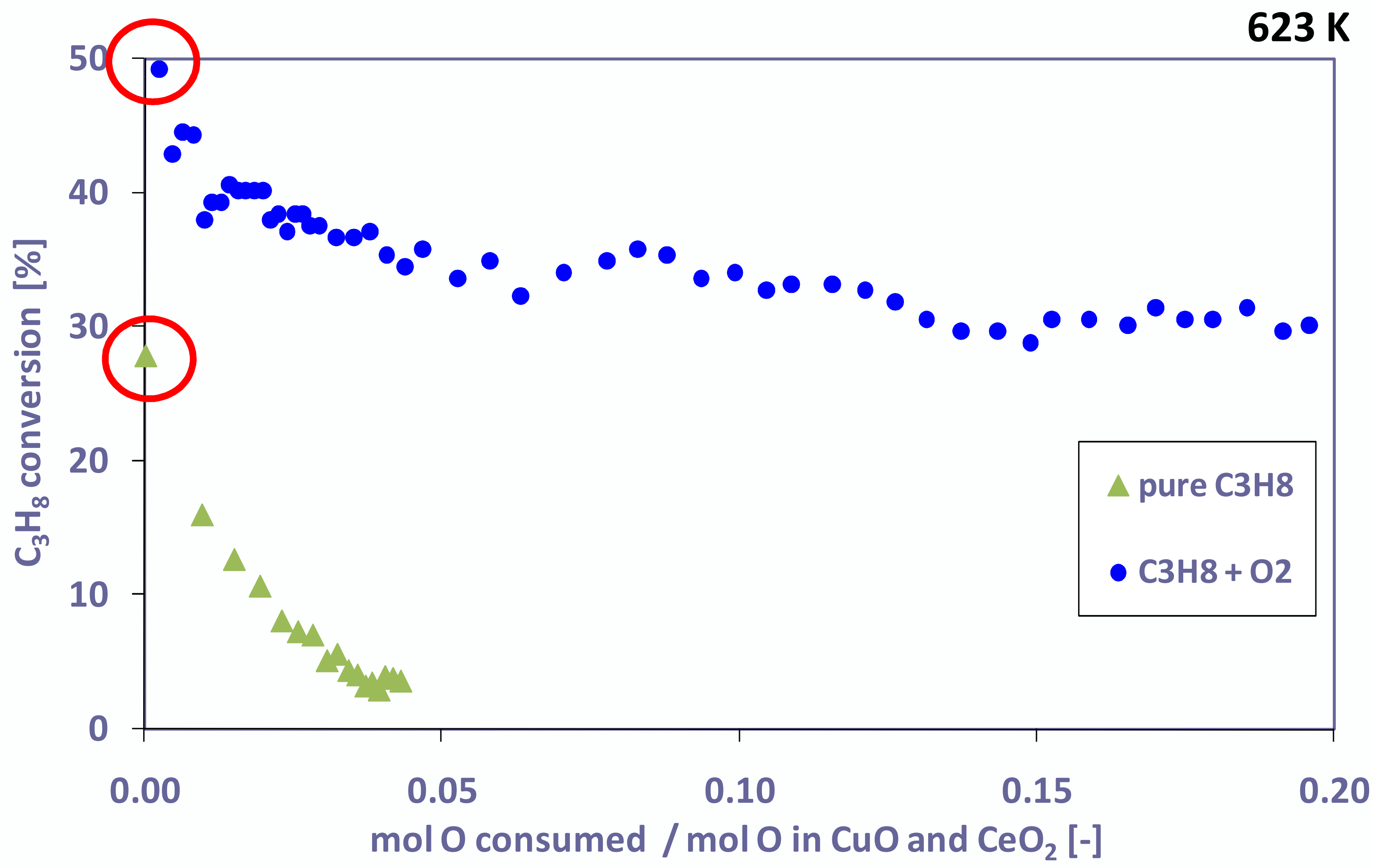
Upper limit for the total number of exchangeable O atoms

pre-treatment of catalyst sample
→ heating to reaction temperature (5K/min)
→ multi-pulses of O_2 until constant level of oxygen reponse

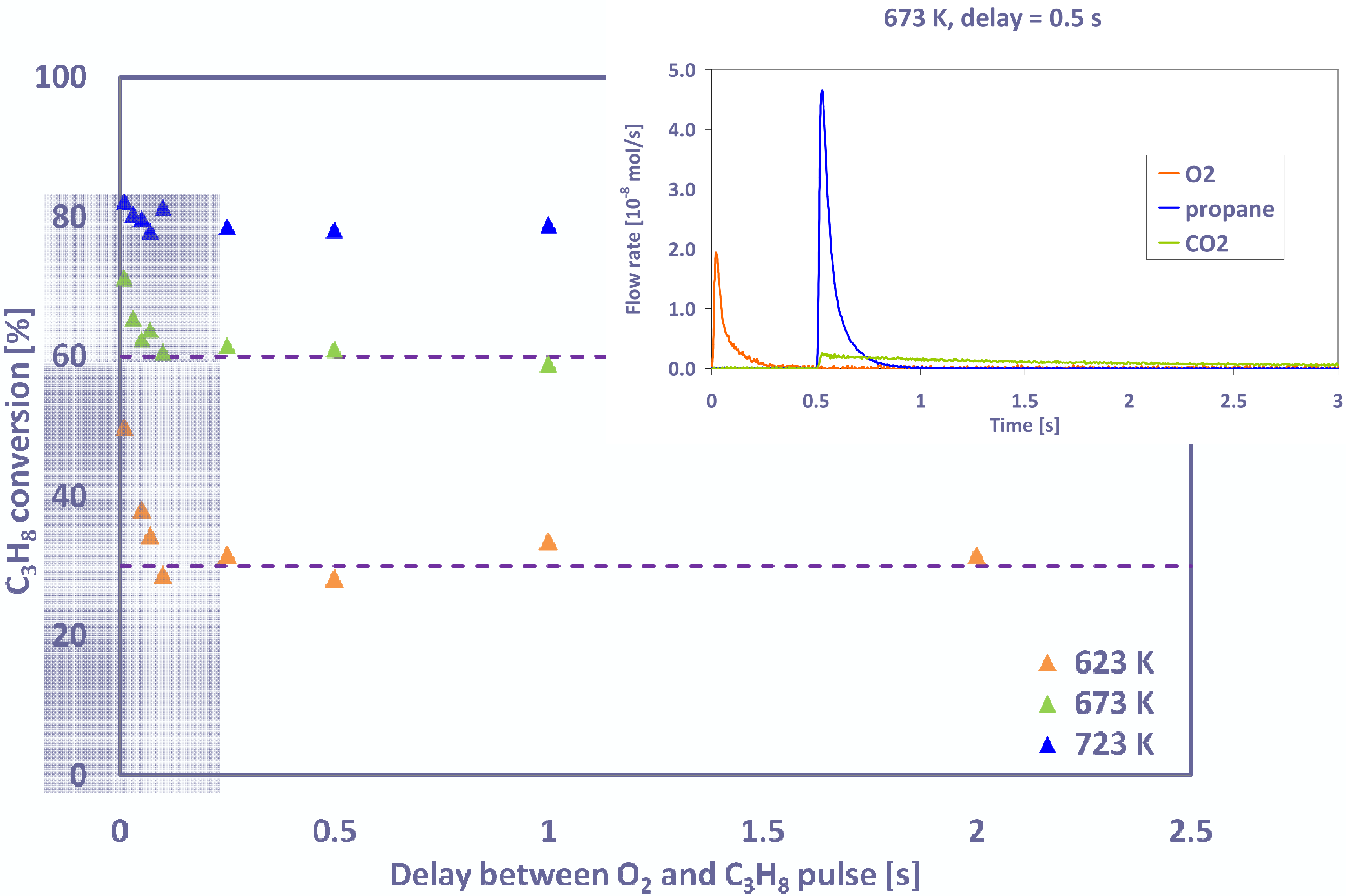
Participation of lattice oxygen at surface



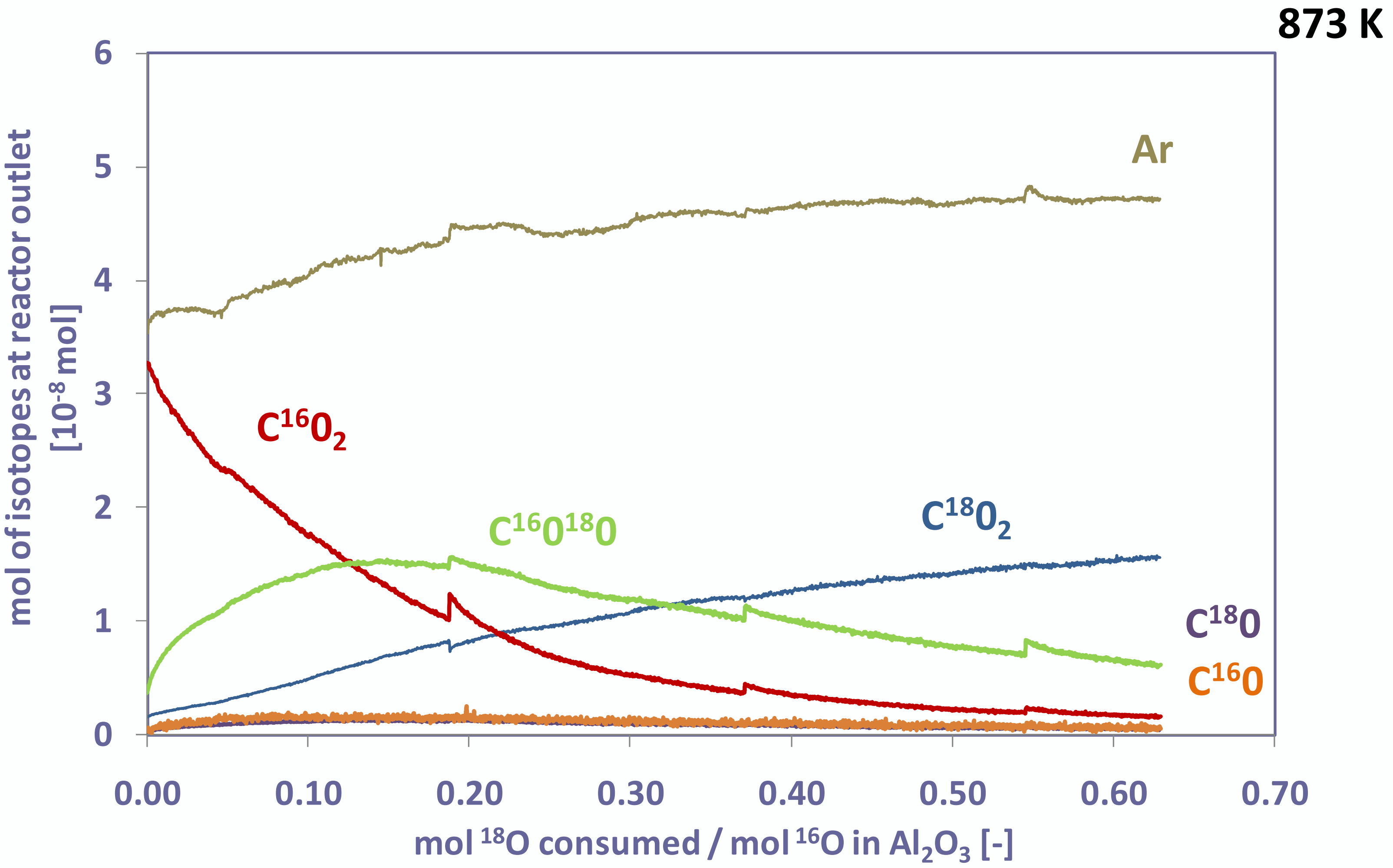
Participation of adsorbed oxygen species



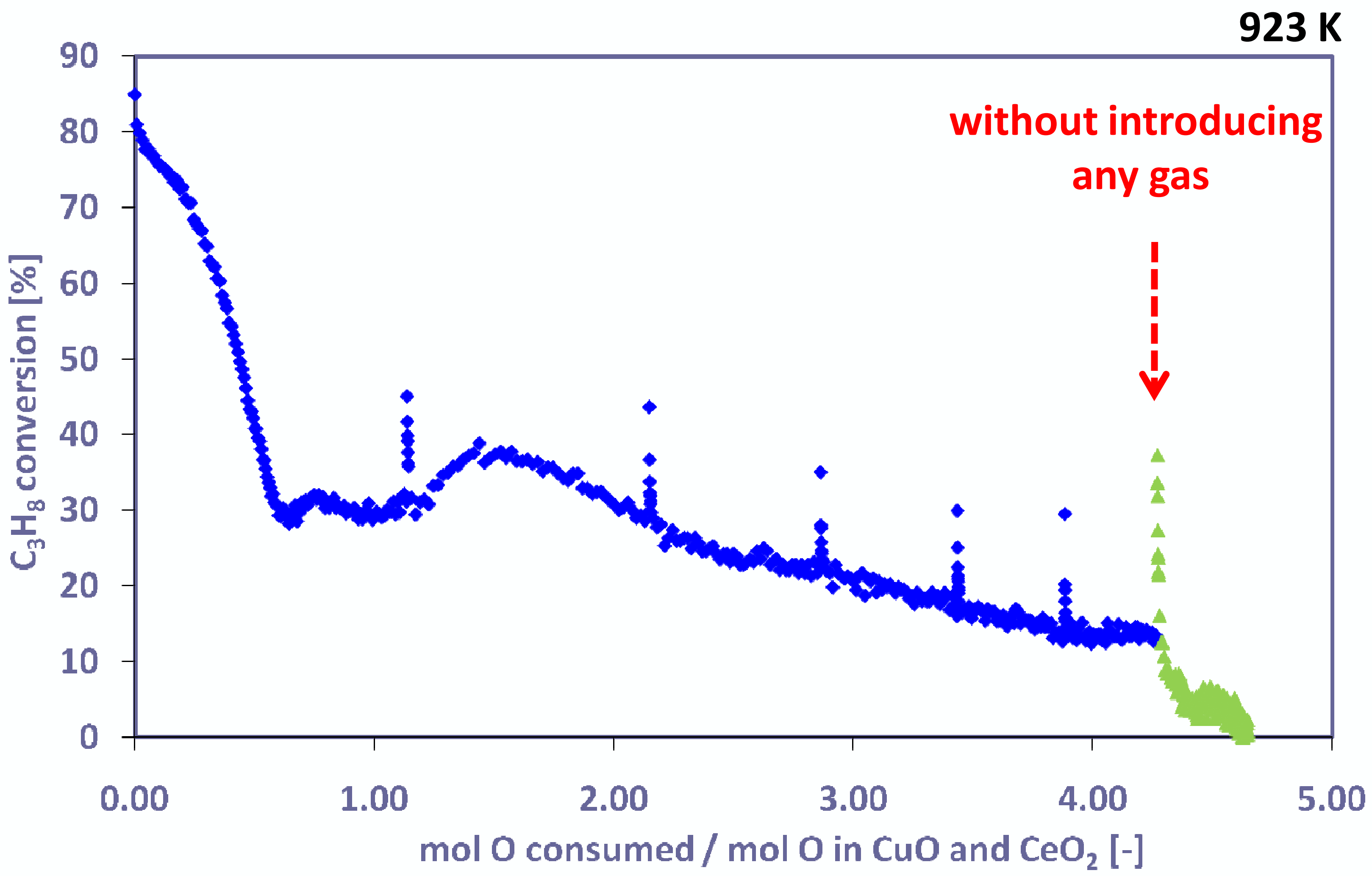
Life time of adsorbed oxygen species



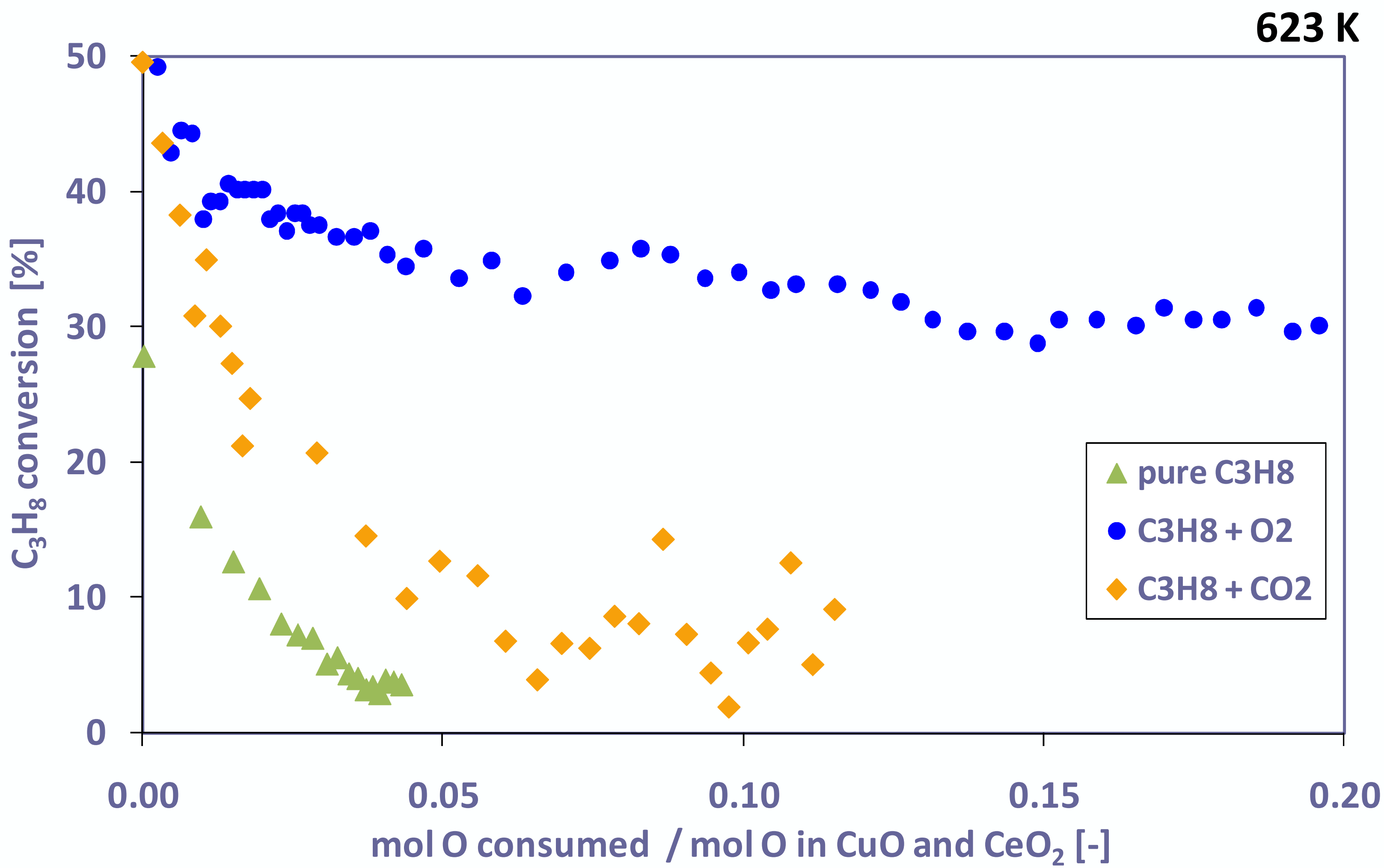
Fast diffusion of oxygen species



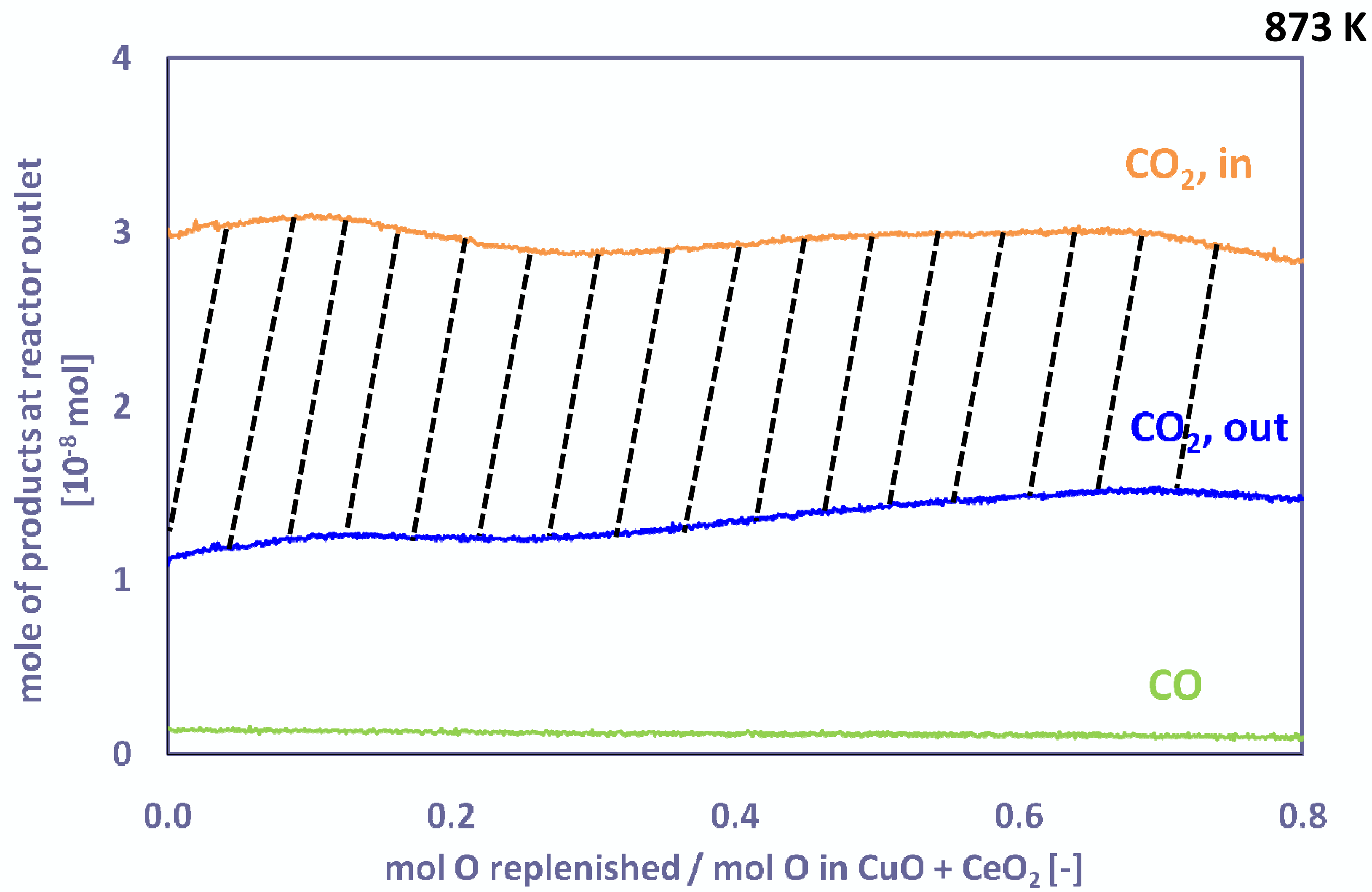
Participation of lattice oxygen from bulk



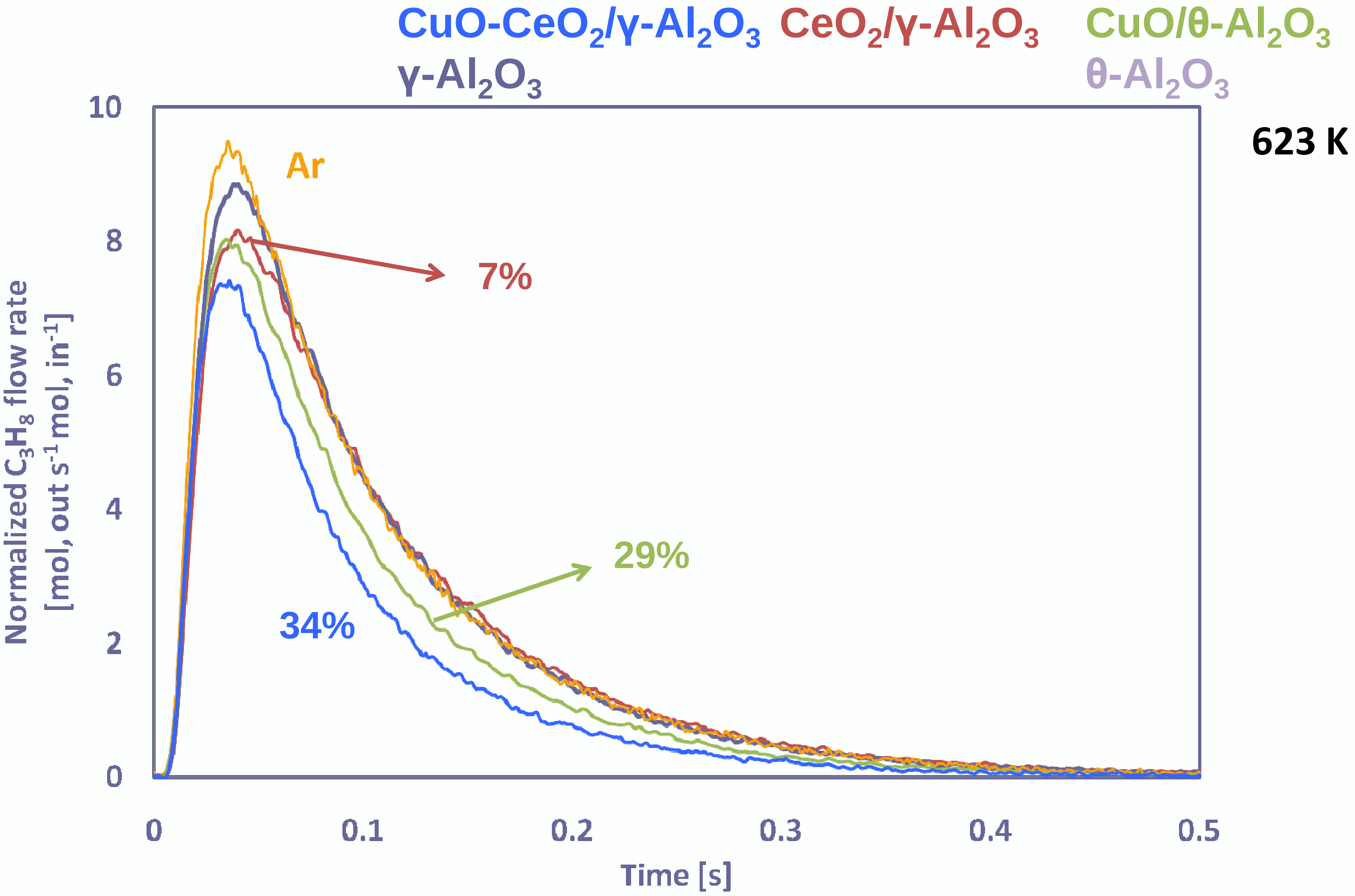
CO_2 as oxidant



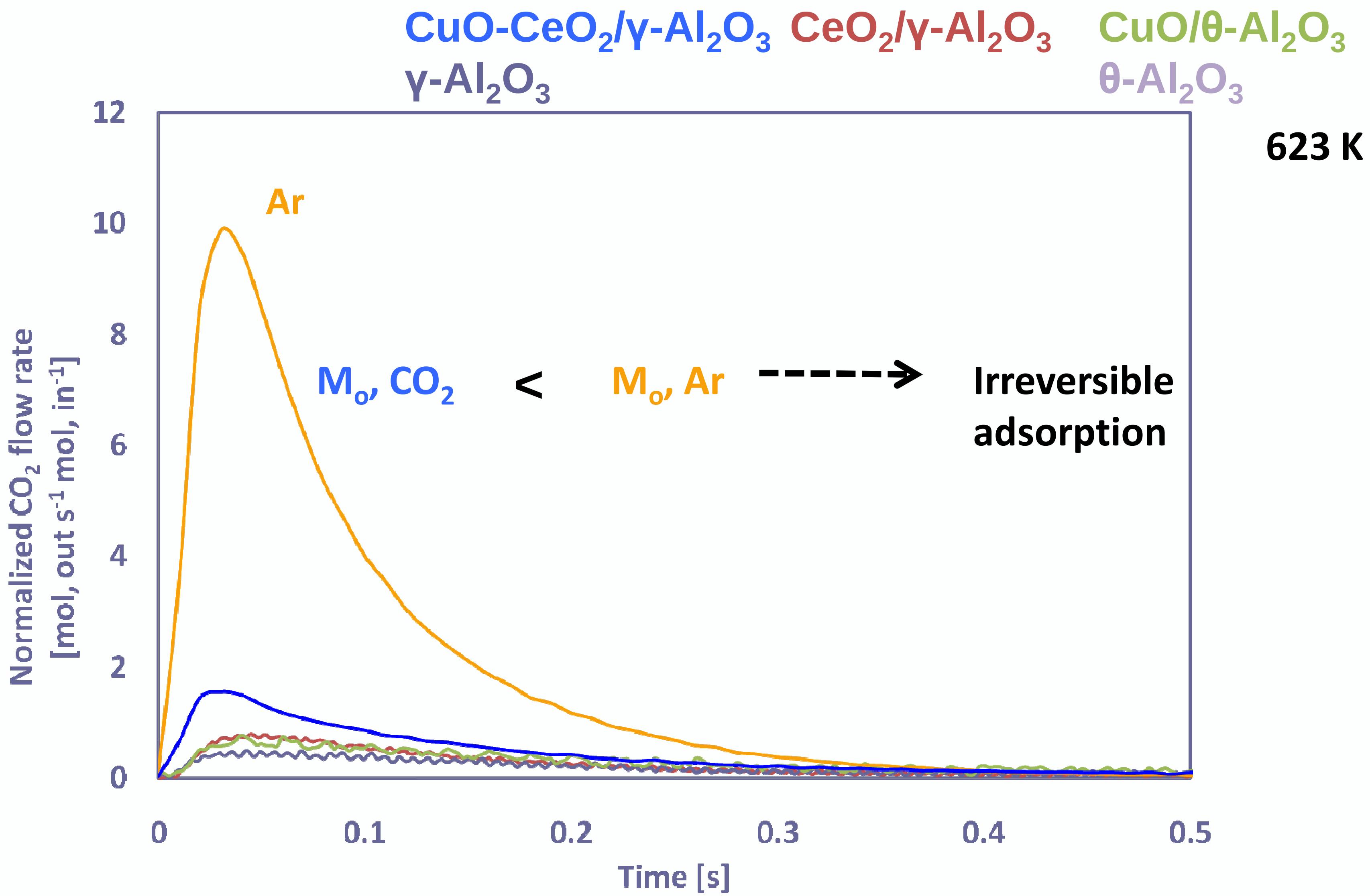
O produced from CO2



Location of active species



CO2 adsorption on alumina



Conclusions

- Four origins of active oxygen species, participating in total oxidation reaction
    1. Lattice oxygen at surface
    2. Lattice oxygen in bulk
    3. Surface oxygen produced from gas-phase O_2
    4. Lattice oxygen produced from gas-phase CO_2
- Location of these active oxygen species
    1. CuO and CeO_2 → active phases → contain active O species
    2. γ-Al_2O_3 → carrier → can produce active O species based on CO_2

This work was performed in the framework of a
Concerted Research Action (Ghent University)

UNIVERSITEIT GENT

Thank you for your attention!

veerle.balcaen@ugent.be

http://www.lct.ugent.be