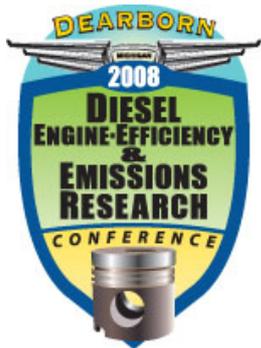




Deactivation Mechanism of Cu/Zelite SCR Catalyst Due to Reductive Hydrothermal Aging

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Introduction

- Cu/zeolite is SCR catalyst candidate for diesel engine
NOx removal
- Cu/zeolite is susceptible to reductive hydrothermal deactivation, it is important to understand failure mode and deactivation mechanism:
 - ≍ DPF regeneration results in O₂ depletion
 - ≍ Aged DOC results in HC deposition on SCR and HC ignition results in O₂ depletion
 - ≍ Engine malfunction causes rich exhaust stream



Catalyst Aging, Testing & Characterization

Reductive Hydrothermal Aging

- Monolithic Cu/zeolite catalyst
- C₃H₆/CO/CO+H₂ presence
- Without O₂ presence
- 5% CO₂, 4.5% H₂O, N₂ Balance
- 30,000/h SV

Activity Testing

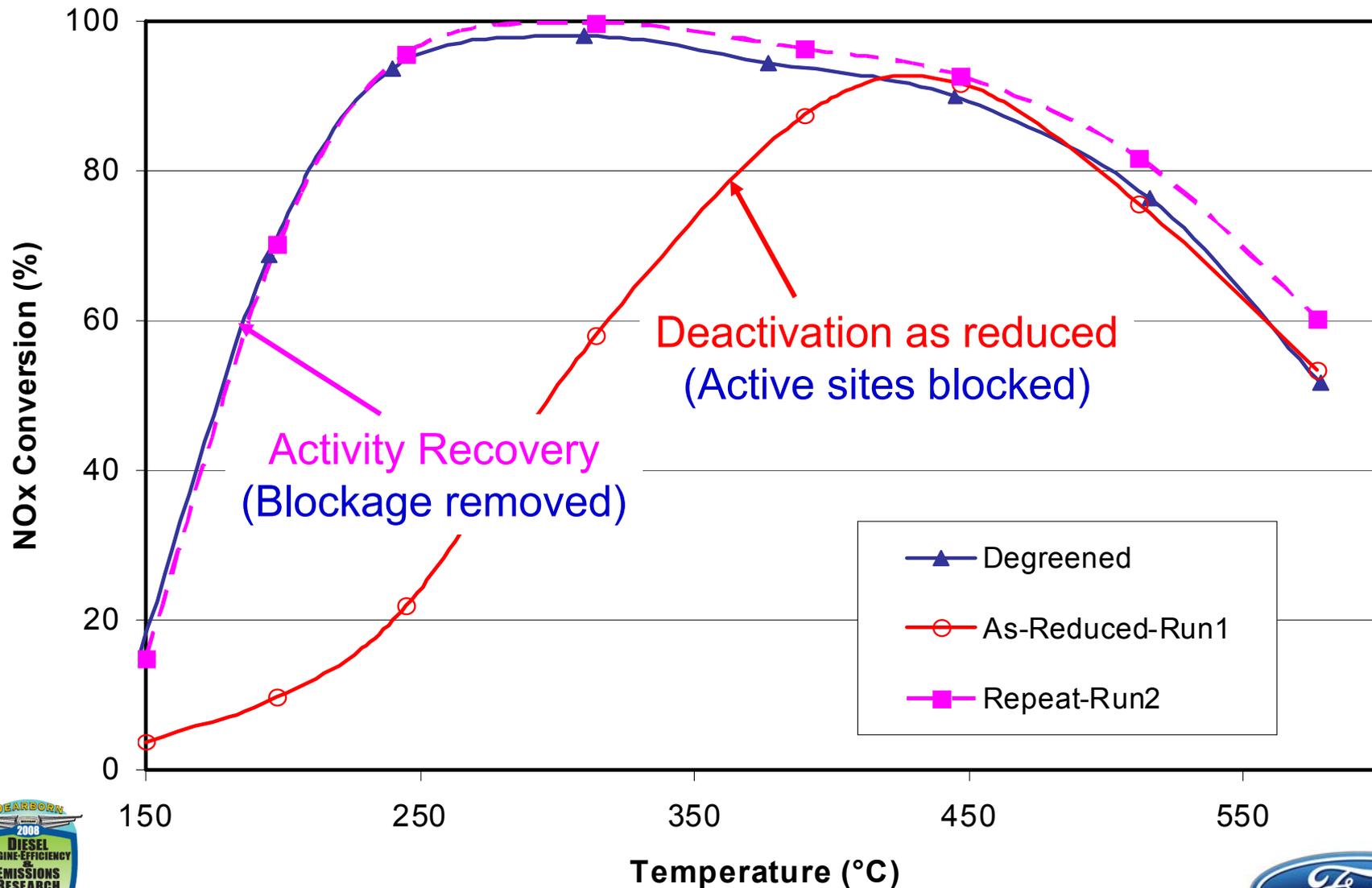
- 350ppm NO
- 350ppm NH₃
- 5% CO₂
- 4.5% H₂O
- N₂ Balance
- 30,000/h SV

Characterization

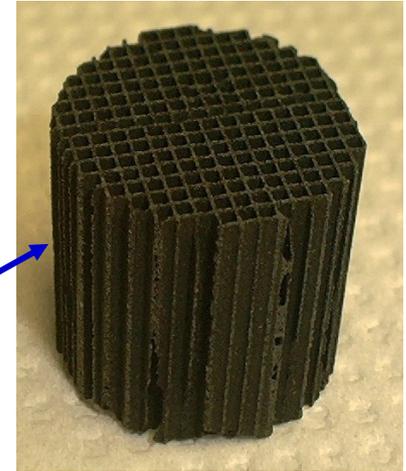
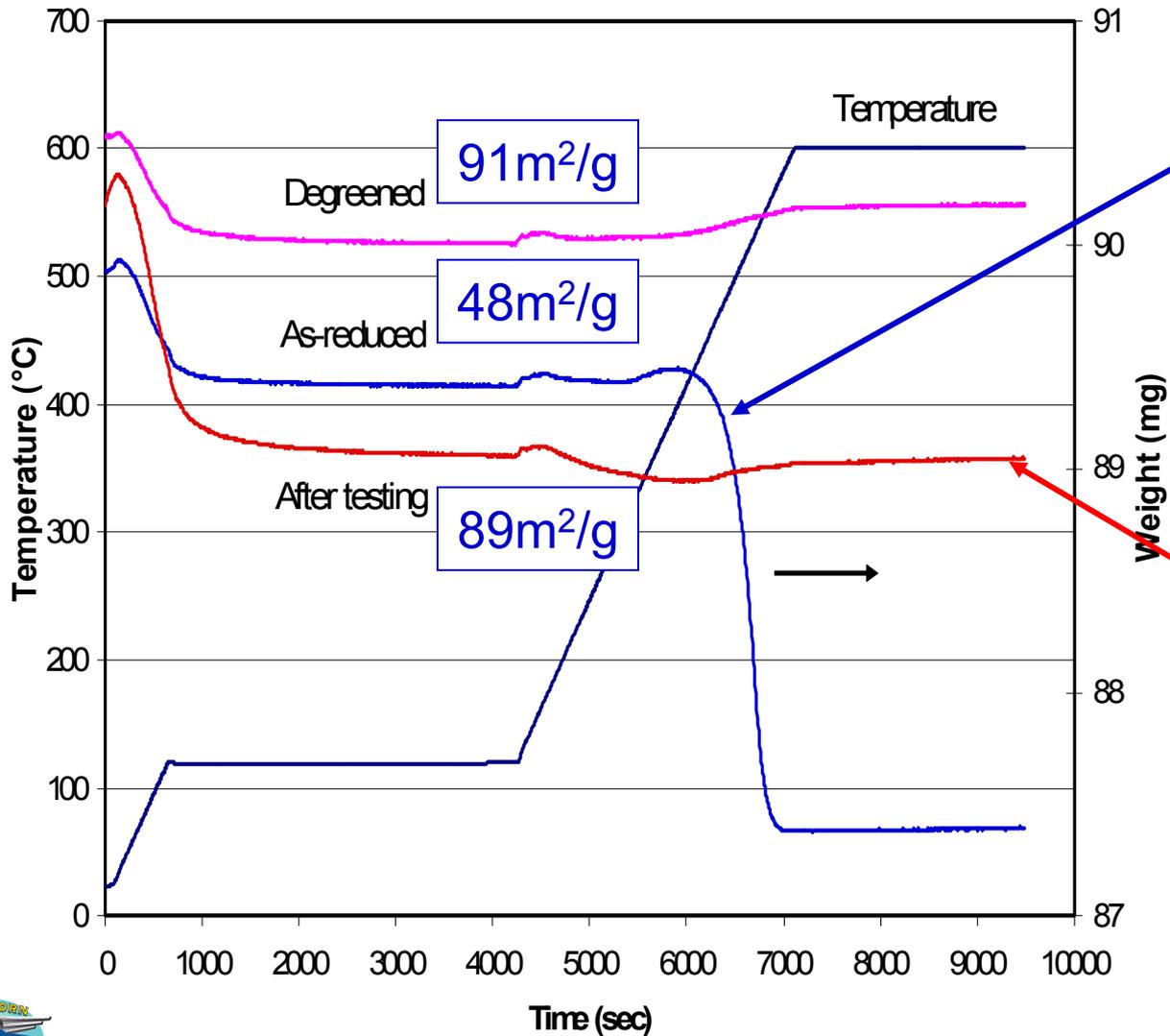
- TGA
- XRD
- BET
- XPS
- N₂ Adsorption

Activities-HC Reduced

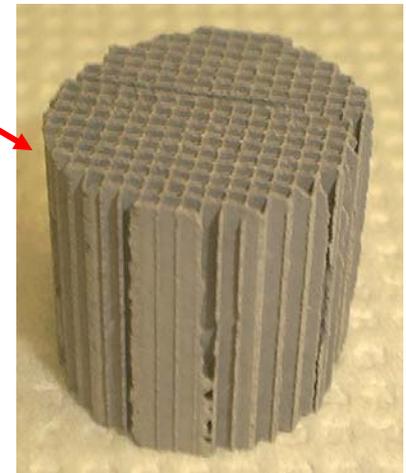
(1000ppm C₃H₆, 0% O₂, 650°C1Hr)



TGA-HC Reduced

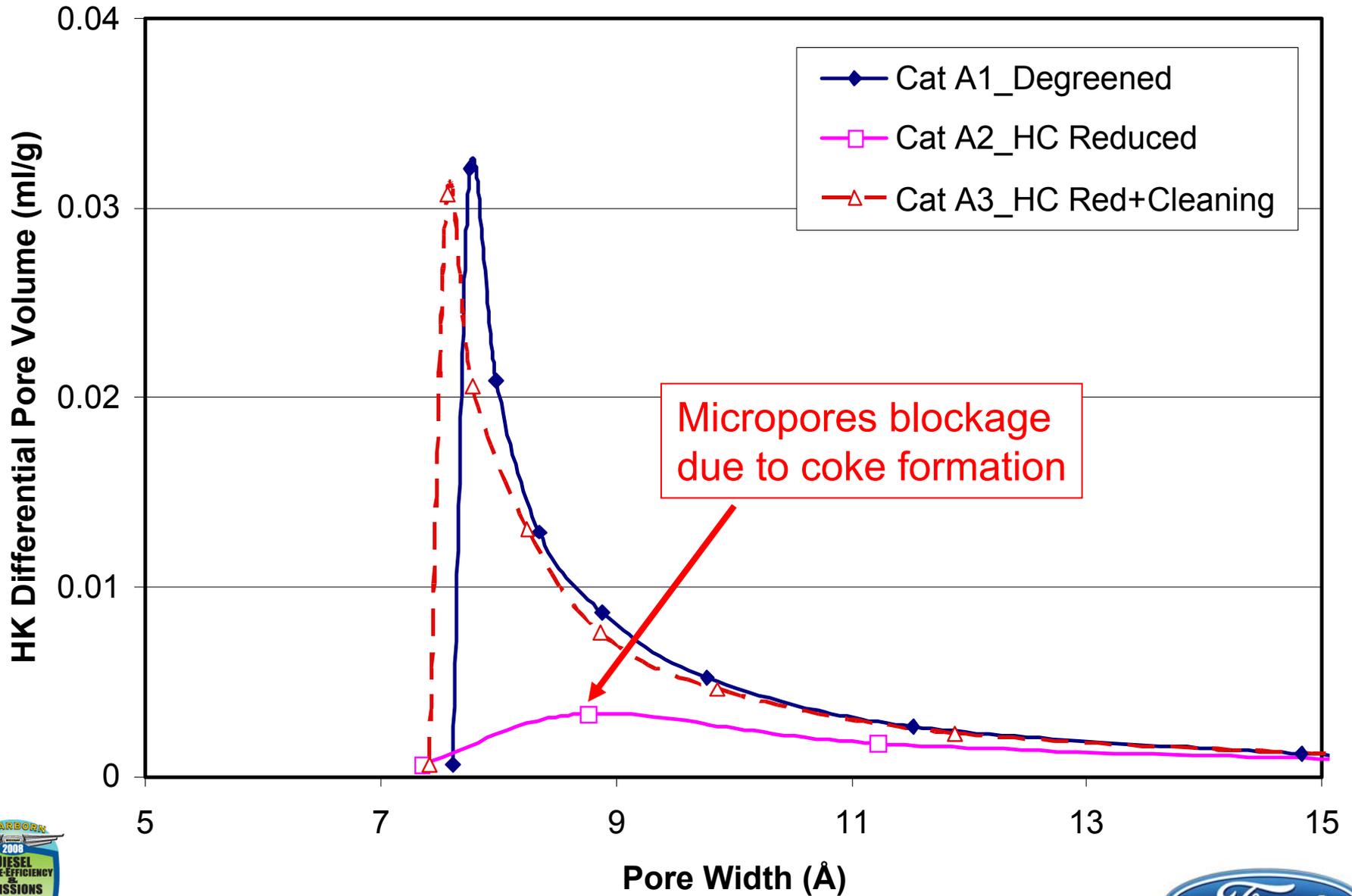


**As-reduced
coke formation**

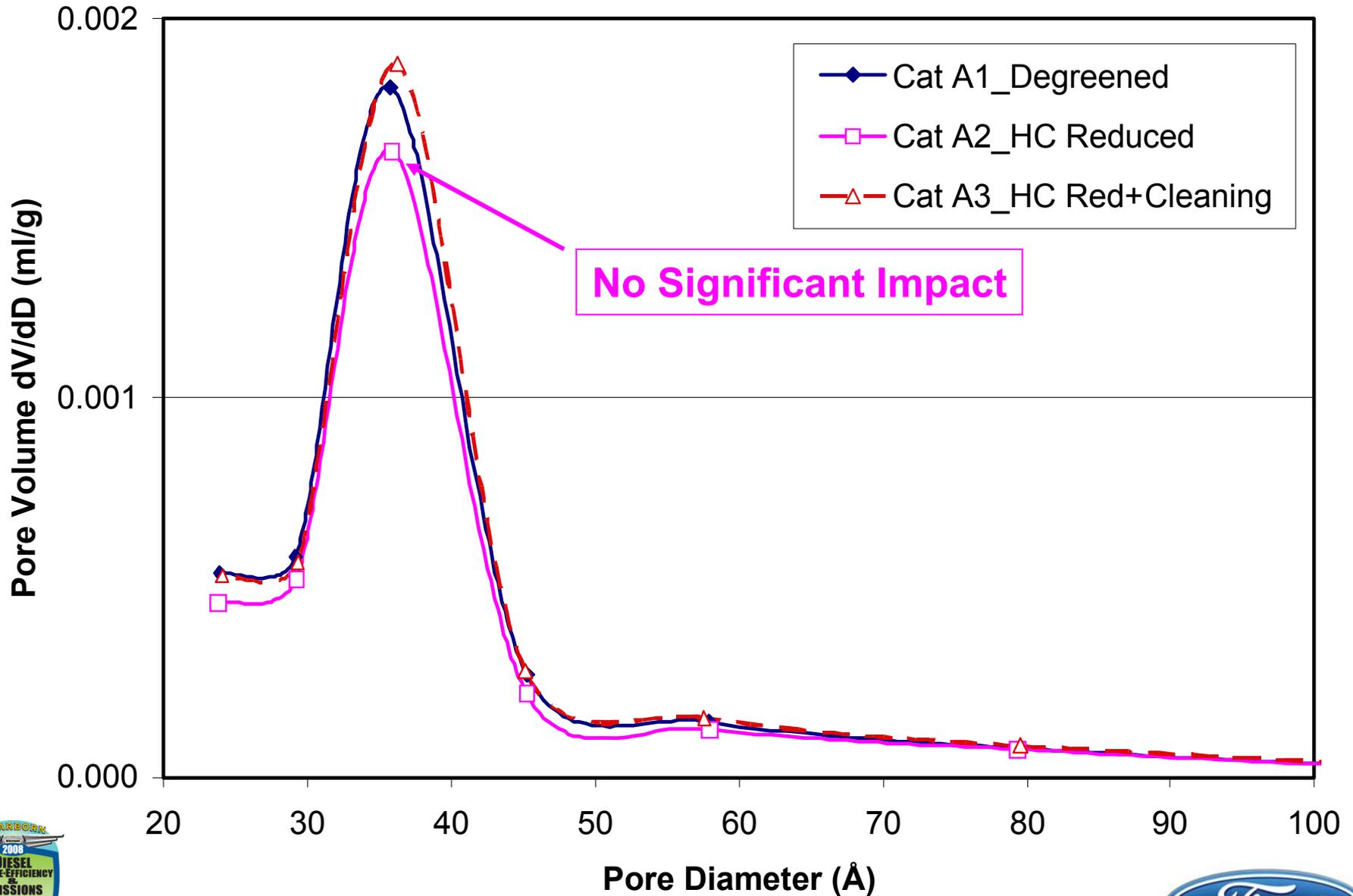


**Coke burnoff
after SCR reaction**

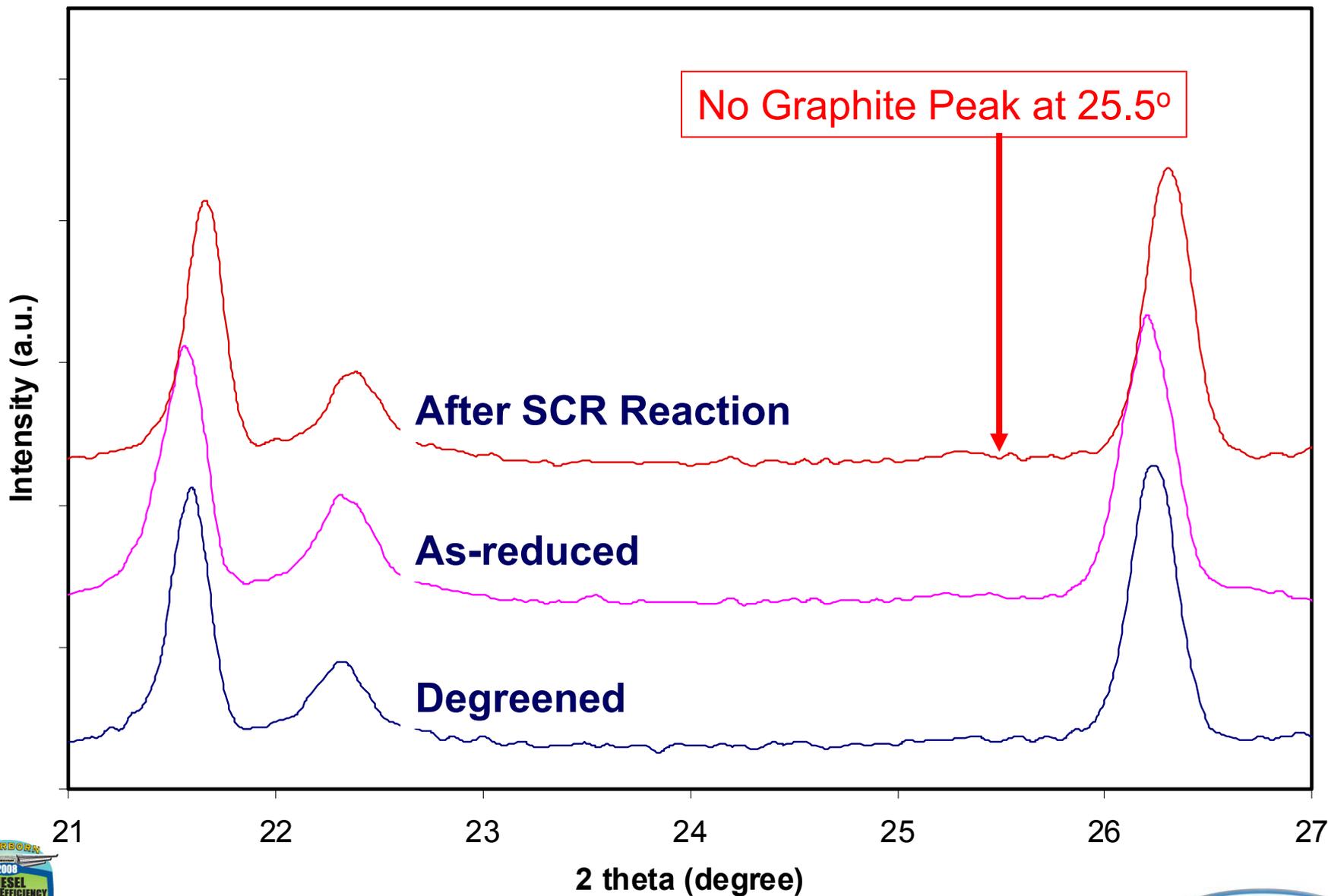
Micropores-HC Reduced



Mesopores-HC Reduced

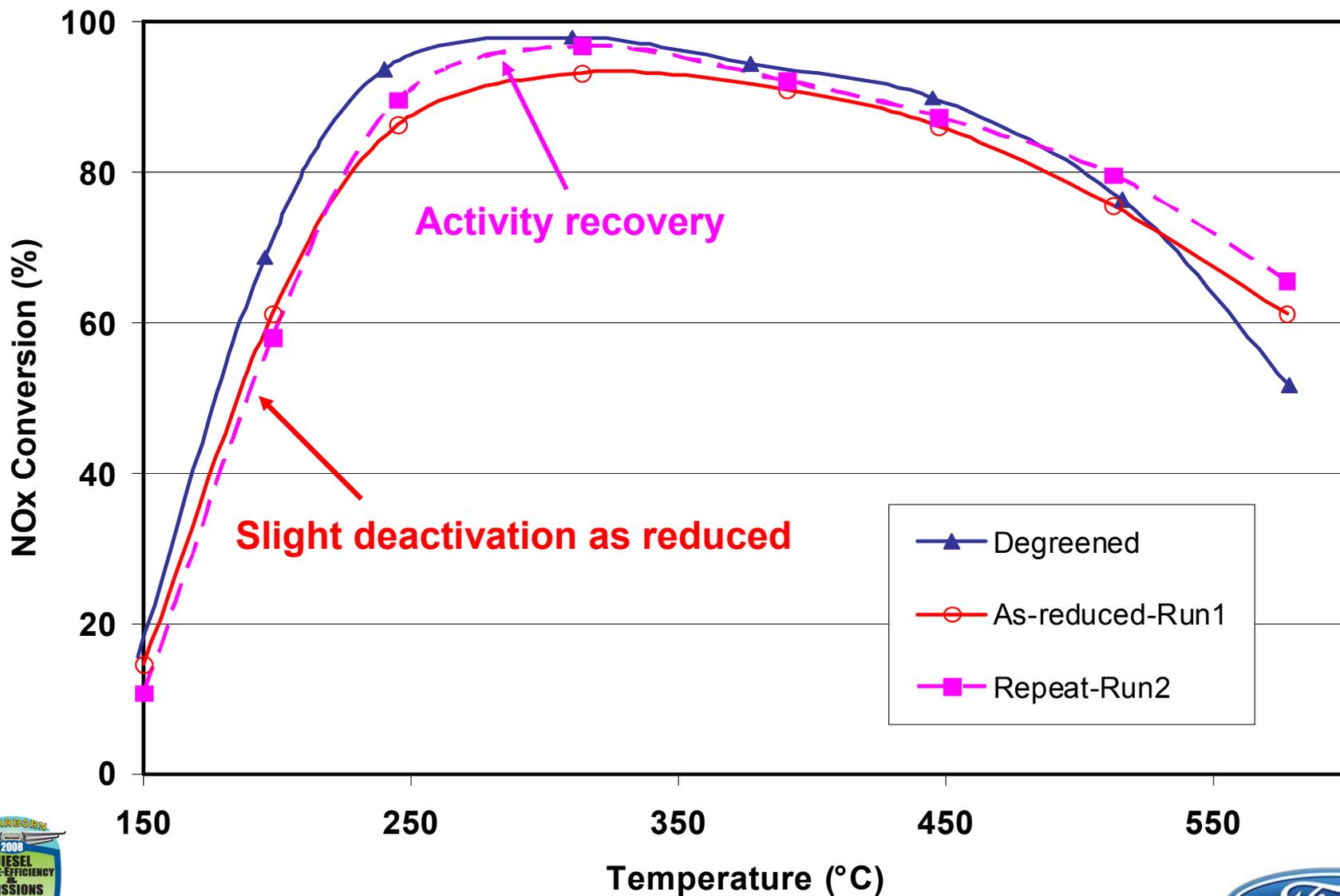


XRD-HC Reduced

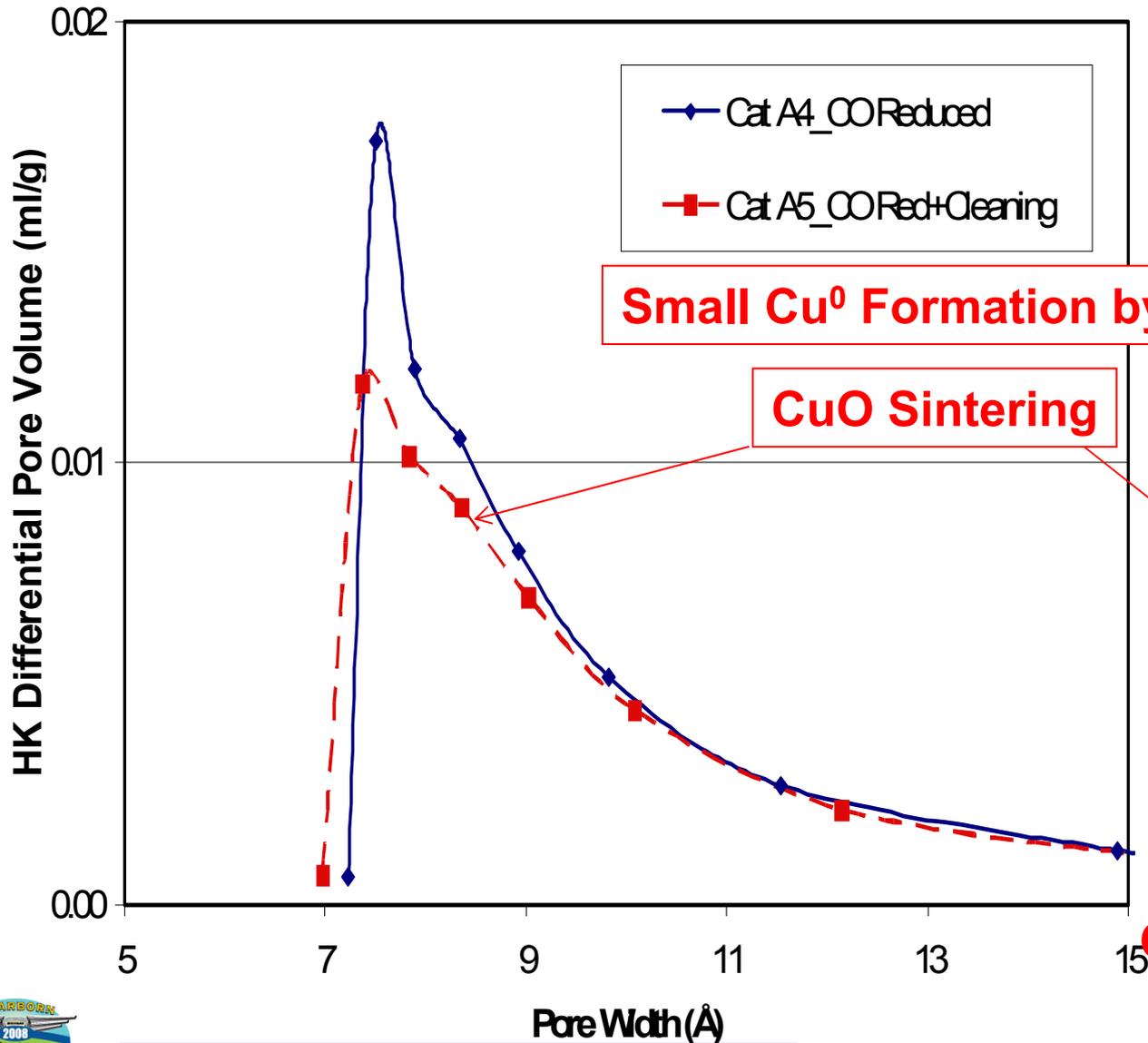


Activities-CO Reduced

(3000ppm CO, 0% O₂, 650°C1Hr)

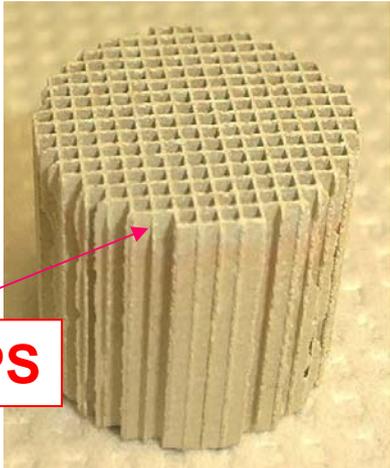


Micropores-CO Reduced

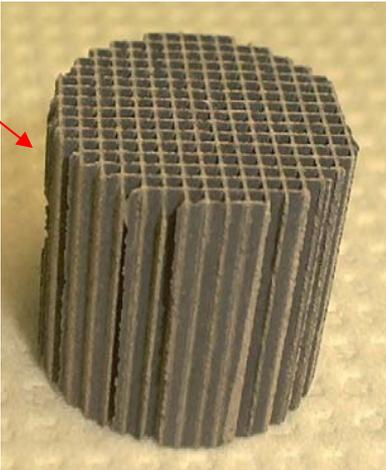


Small Cu⁰ Formation by XPS

CuO Sintering



As-reduced

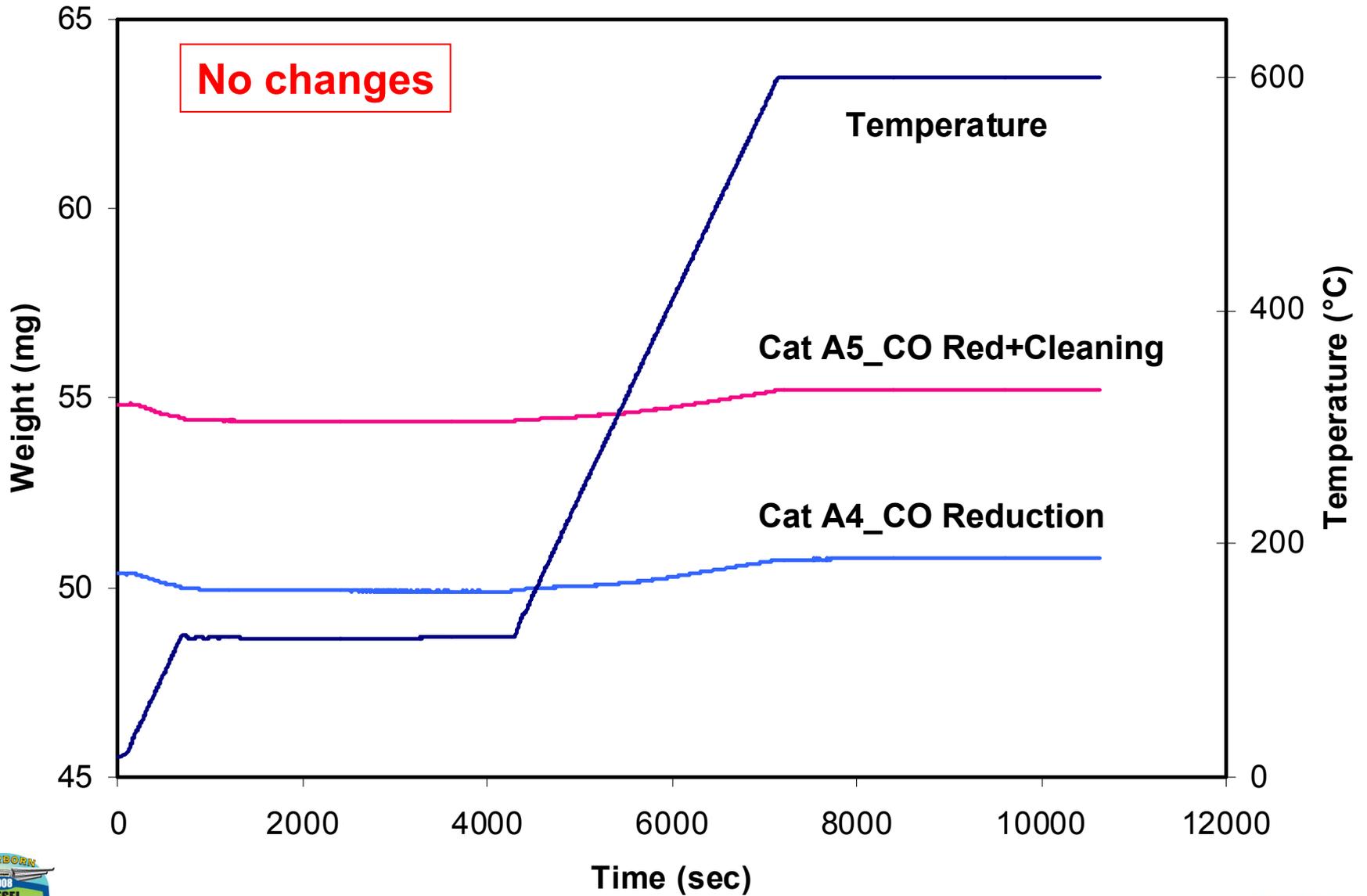


CO Red.+SCR reaction

Mesopores have no change

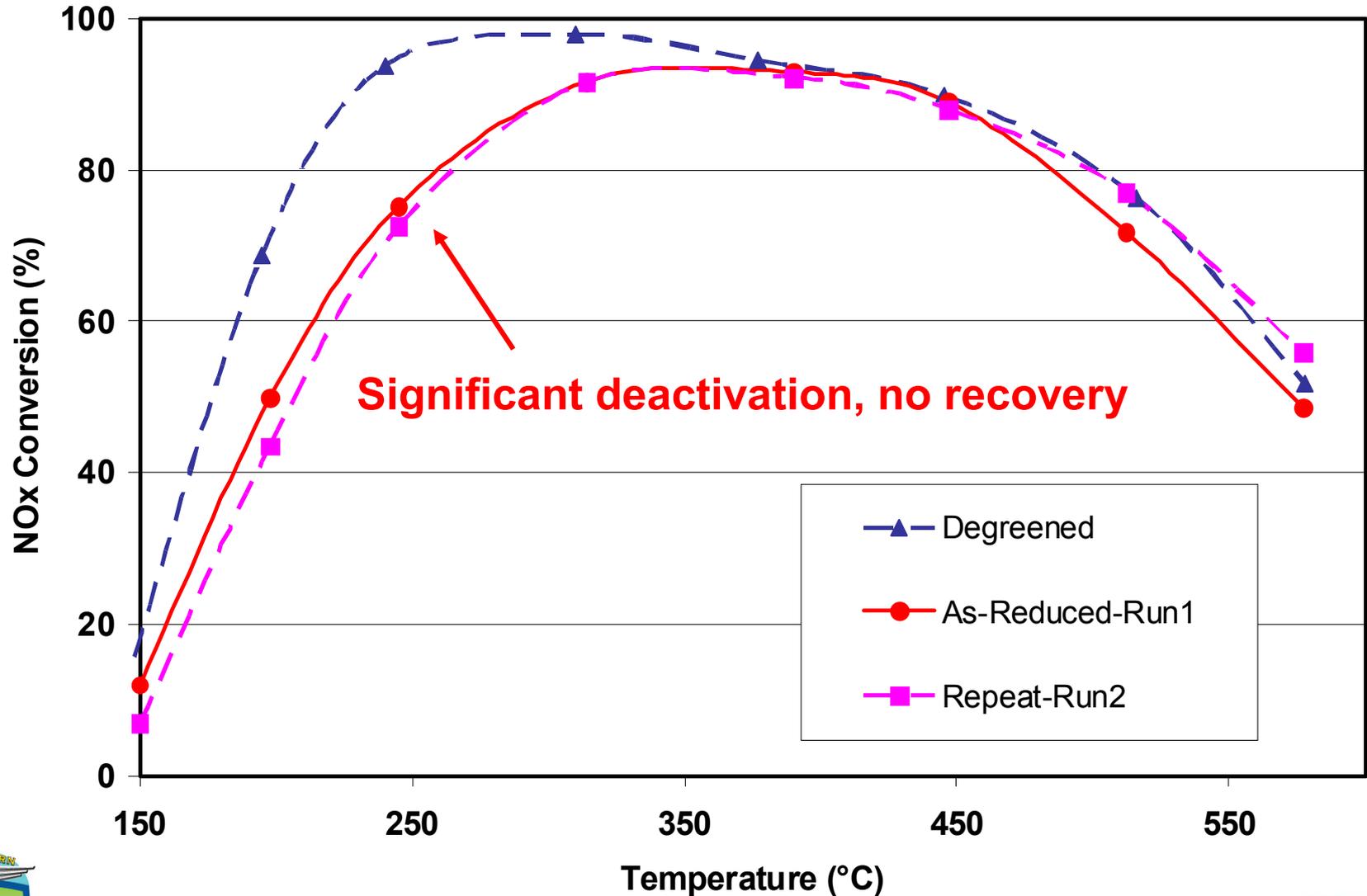


TGA-CO Reduced

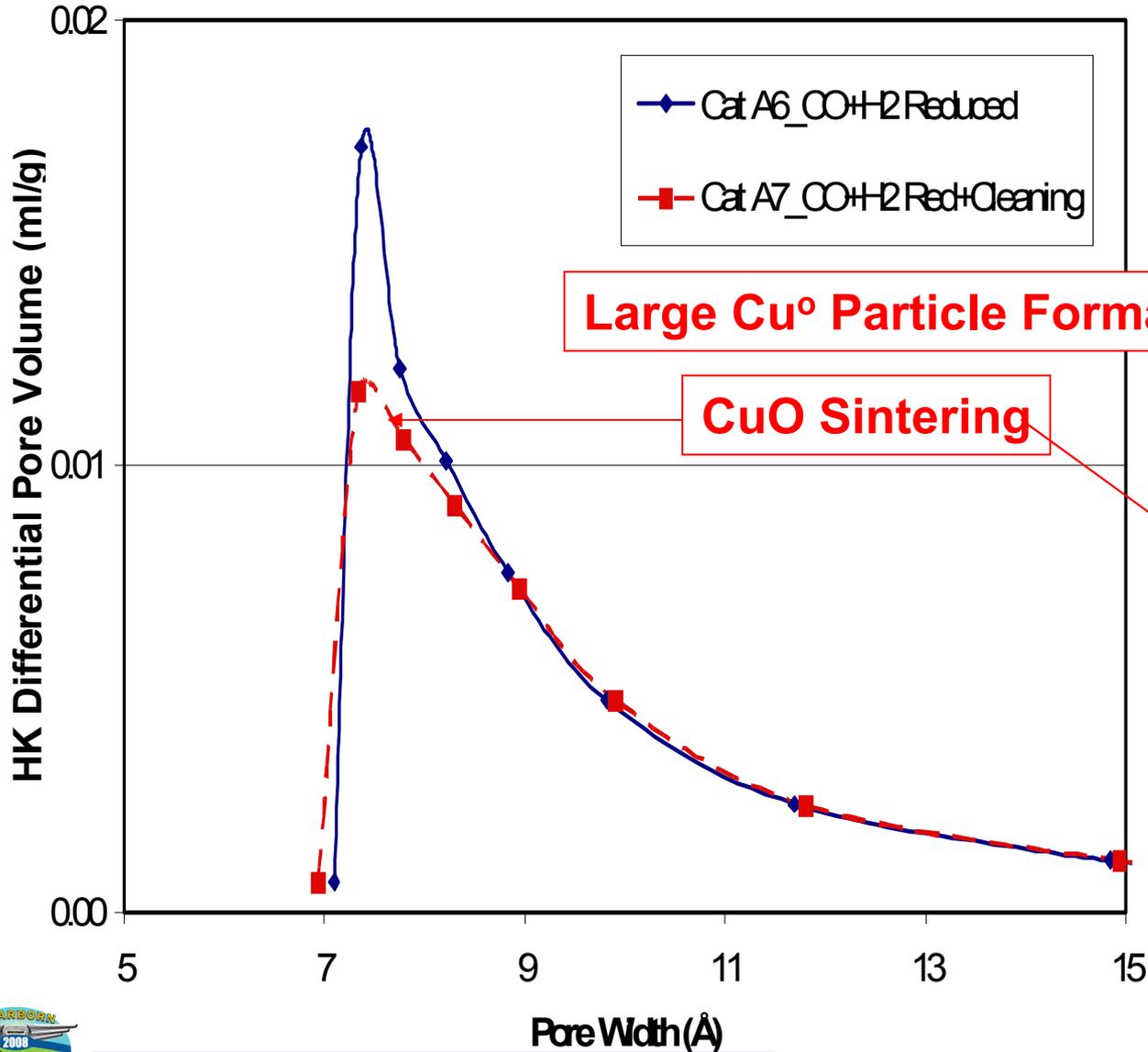


Activities-CO+H₂ Reduced

(3000ppm CO + 1000ppm H₂, 0% O₂, 650°C, 1hr)

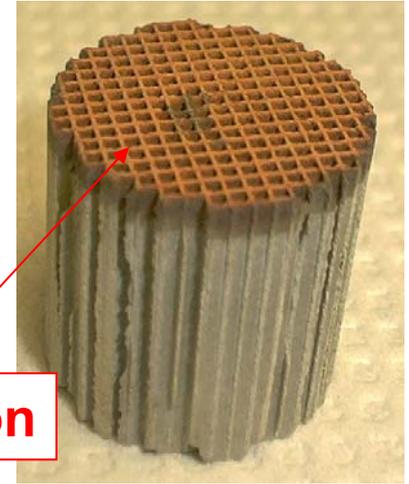


Micropores-CO+H₂ Reduced

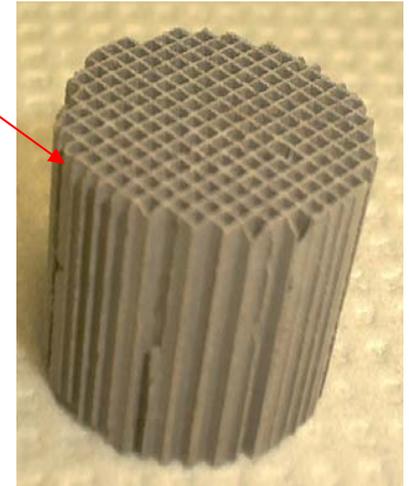


Large Cu⁰ Particle Formation

CuO Sintering



As-reduced

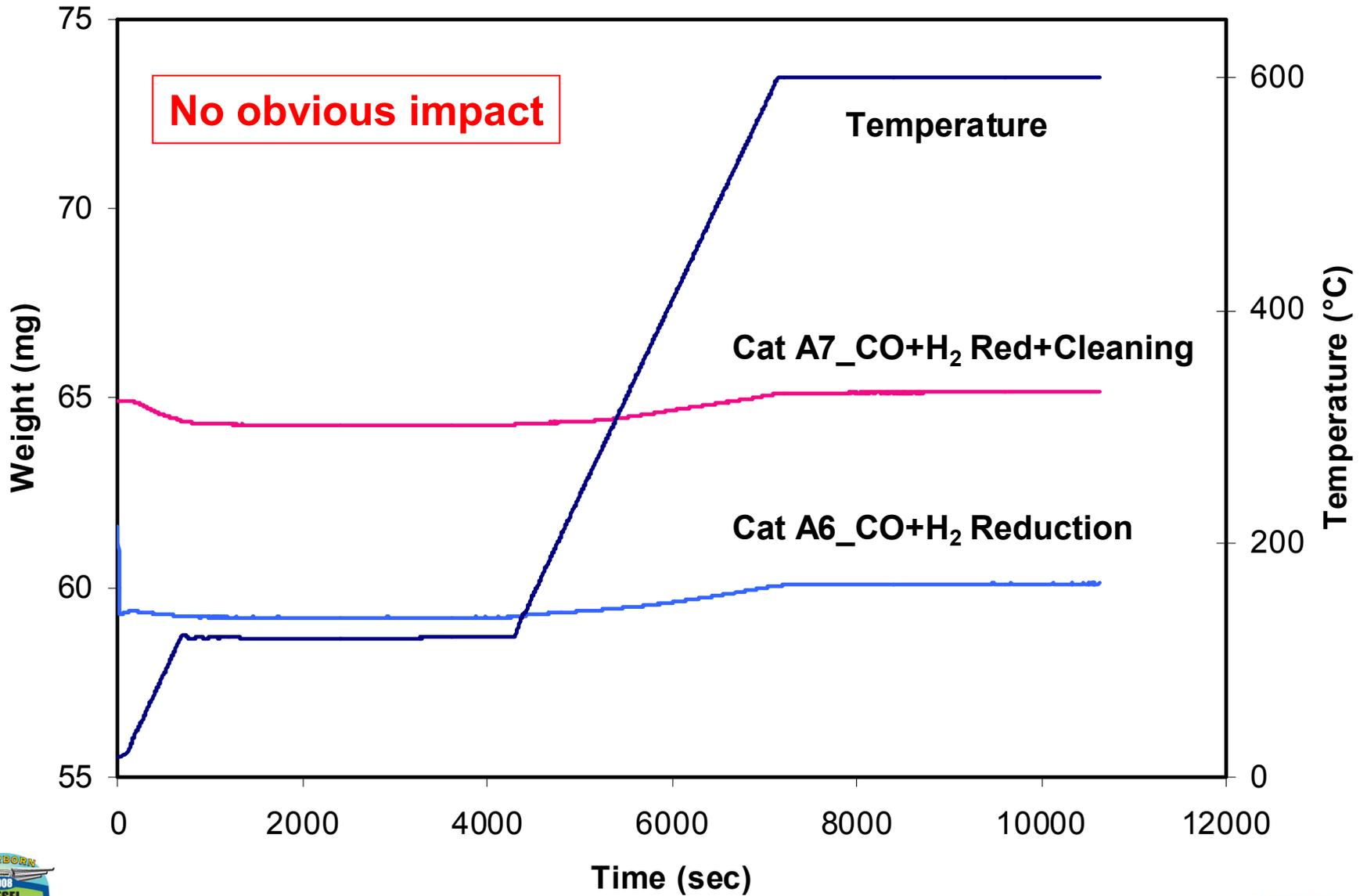


CO+H₂ Reduction +SCR reaction

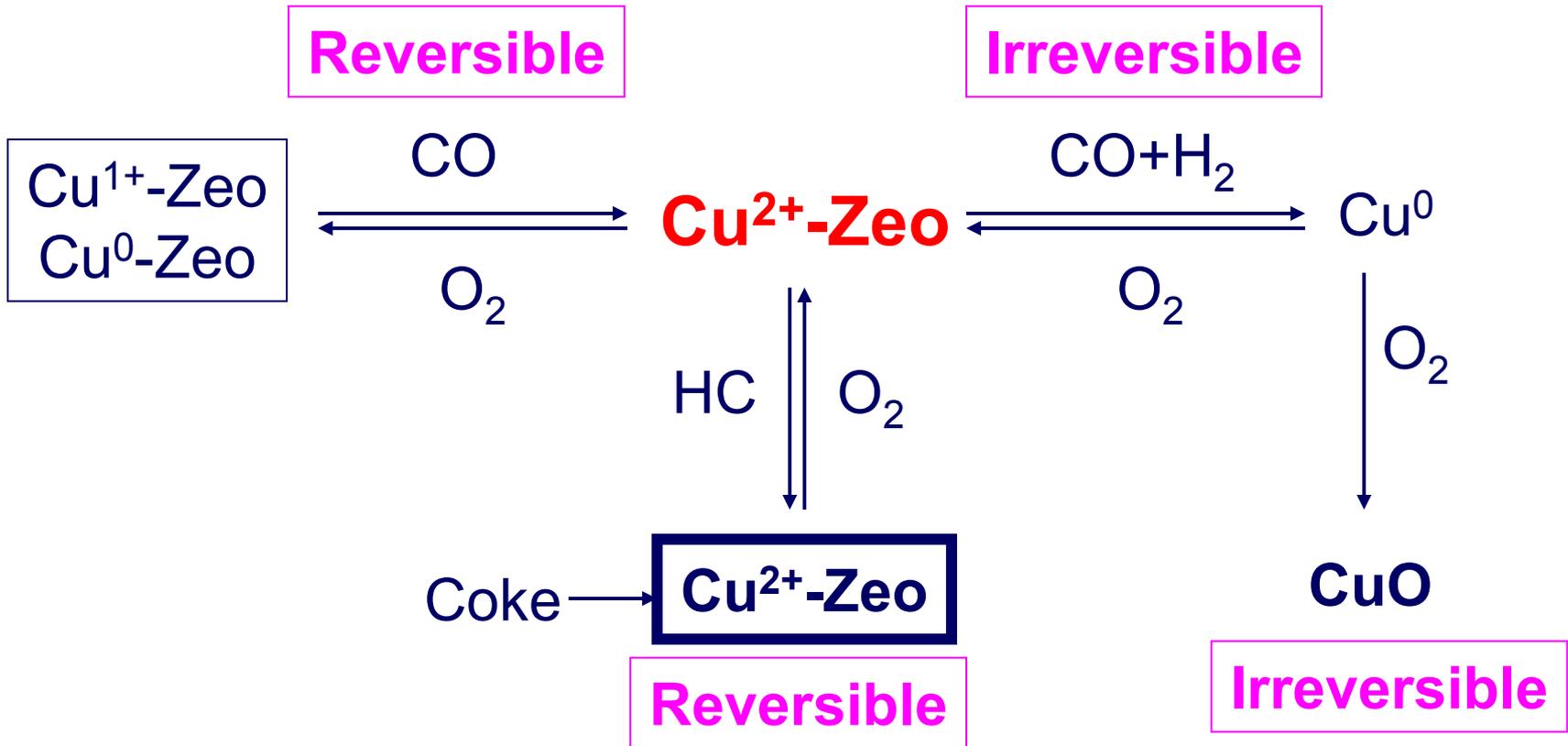
Mesopores have no change



TGA-CO+H₂ Reduced



Chemistry of Cu/Zeolite Catalyst



Deactivation: Cu^{2+} reduction – Cu oxidation – CuO sintering

Summary

- HC reduction results in coke formation and reversible catalyst deactivation
- CO/H₂ reduction results in permanent catalyst deactivation
- Cu²⁺ reduction and sintering are attributed to catalyst deactivation



Thanks!

