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## **Lean NO<sub>x</sub> removal by a bifunctional (EtOH+NH<sub>3</sub>) mixture dedicated to (Ag/Al<sub>2</sub>O<sub>3</sub>+NH<sub>3</sub>-SCR) dual-bed catalytic system.**

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# Lean NO<sub>x</sub> removal by a bifunctional (EtOH+NH<sub>3</sub>) mixture dedicated to (Ag/Al<sub>2</sub>O<sub>3</sub>+NH<sub>3</sub>-SCR) dual-bed catalytic system. Comparison between WO<sub>3</sub>/CeZrO<sub>2</sub> and Cu/FER as NH<sub>3</sub>-SCR catalyst.

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## INTRODUCTION low efficiency of the NO SCR (by NH<sub>3</sub>, EtOH...) at T < 250°C

### EtOH-SCR over 2%Ag/Al<sub>2</sub>O<sub>3</sub>

#### Between 175 and 250°C (low T)

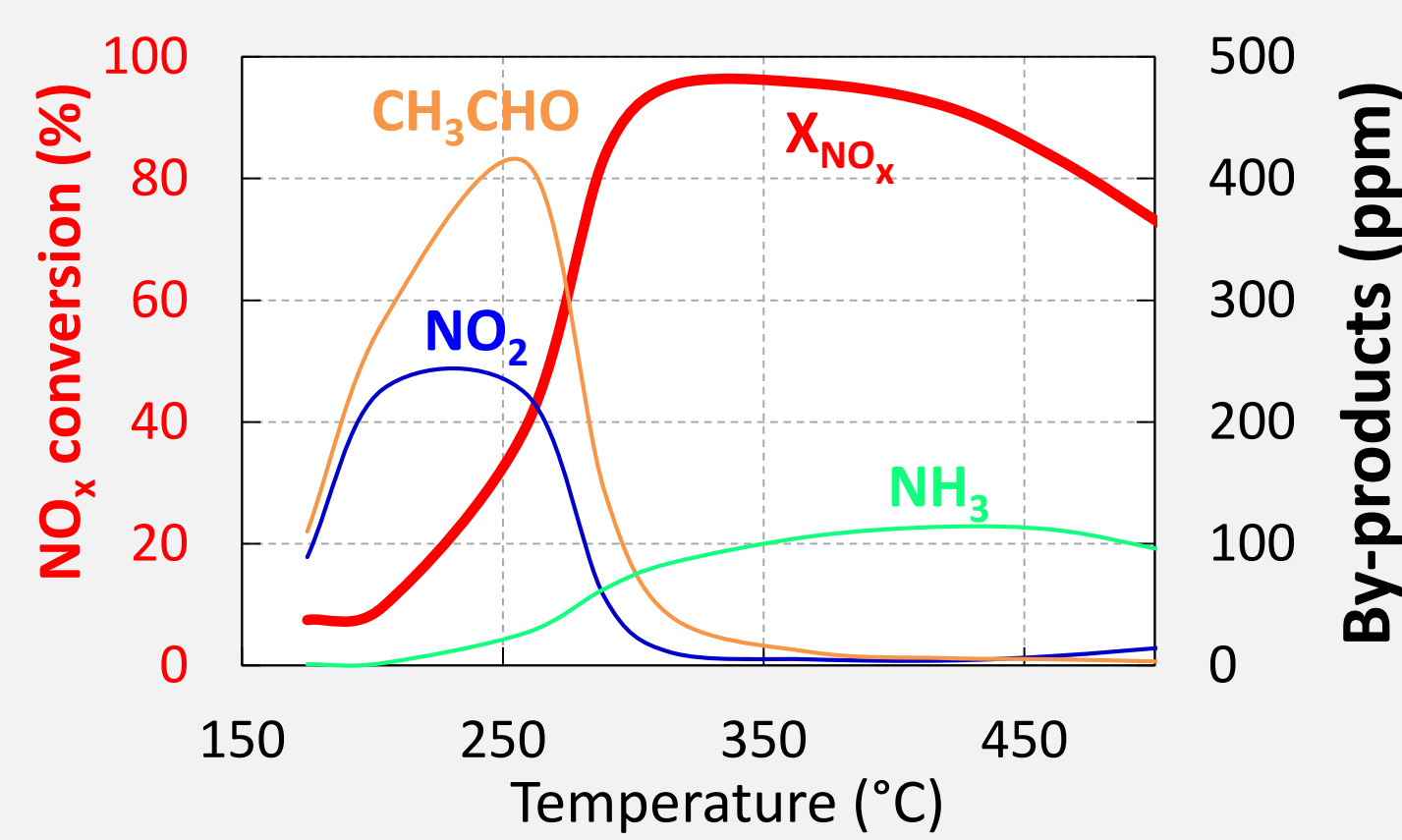
- High NO<sub>2</sub> release (only NO initially)
- Low NO<sub>x</sub> conversion

#### Between 250 and 500°C

- NO<sub>2</sub> and CH<sub>3</sub>CHO react to form reductive C-N ad-species
- NH<sub>3</sub> released while NO<sub>x</sub> conversion increases

→ Is NH<sub>3</sub> formation the limiting step of the SCR at low temperature?

**Objectives:** Increase deNO<sub>x</sub> activity at low T by (i) co-feeding of ammonia and ethanol



## Experimental conditions

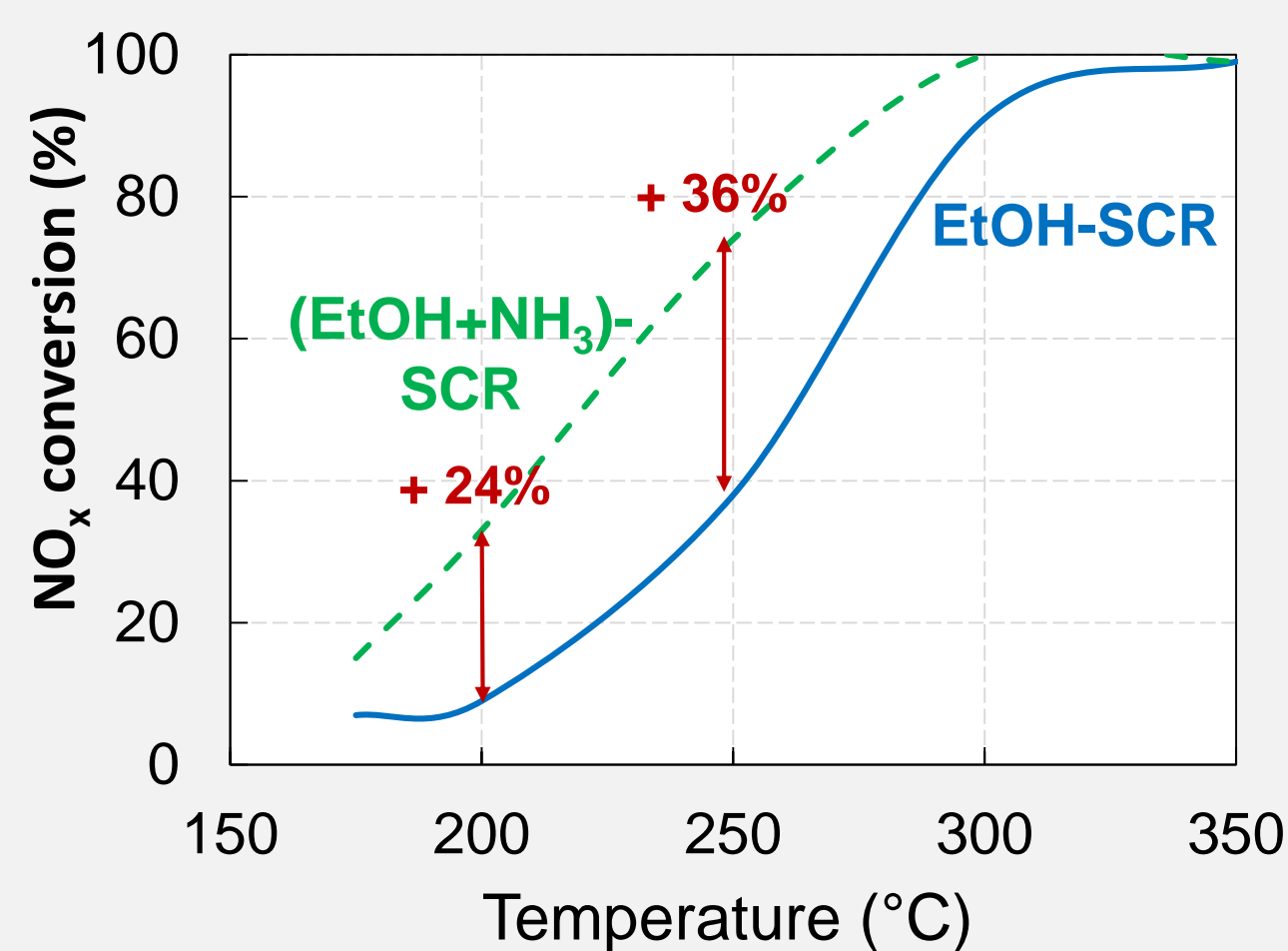
- Catalysts:**
- 2 % Ag/Al<sub>2</sub>O<sub>3</sub>
  - 6 % WO<sub>3</sub>/Ce-Zr-O<sub>2</sub> (40%Ce-60%Zr) aged at 700°C during 4 h under wet air
  - 2.5 % Cu/FER (SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> = 20) aged at 600°C during 16 h under wet air

**Catalytic tests:**

Test	EtOH (ppm)	NH <sub>3</sub> (ppm)	NO (ppm)	NO <sub>2</sub> (ppm)	Additional gases
EtOH-SCR	1200	-	400	-	10% O <sub>2</sub>
(EtOH+NH <sub>3</sub> )-SCR	1200	400	400	-	10% CO <sub>2</sub>
Std NH <sub>3</sub> -SCR	-	400	400	-	8% H <sub>2</sub> O
Fast NH <sub>3</sub> -SCR	-	400	200	200	N <sub>2</sub>

GHSV = 130 000 h<sup>-1</sup>

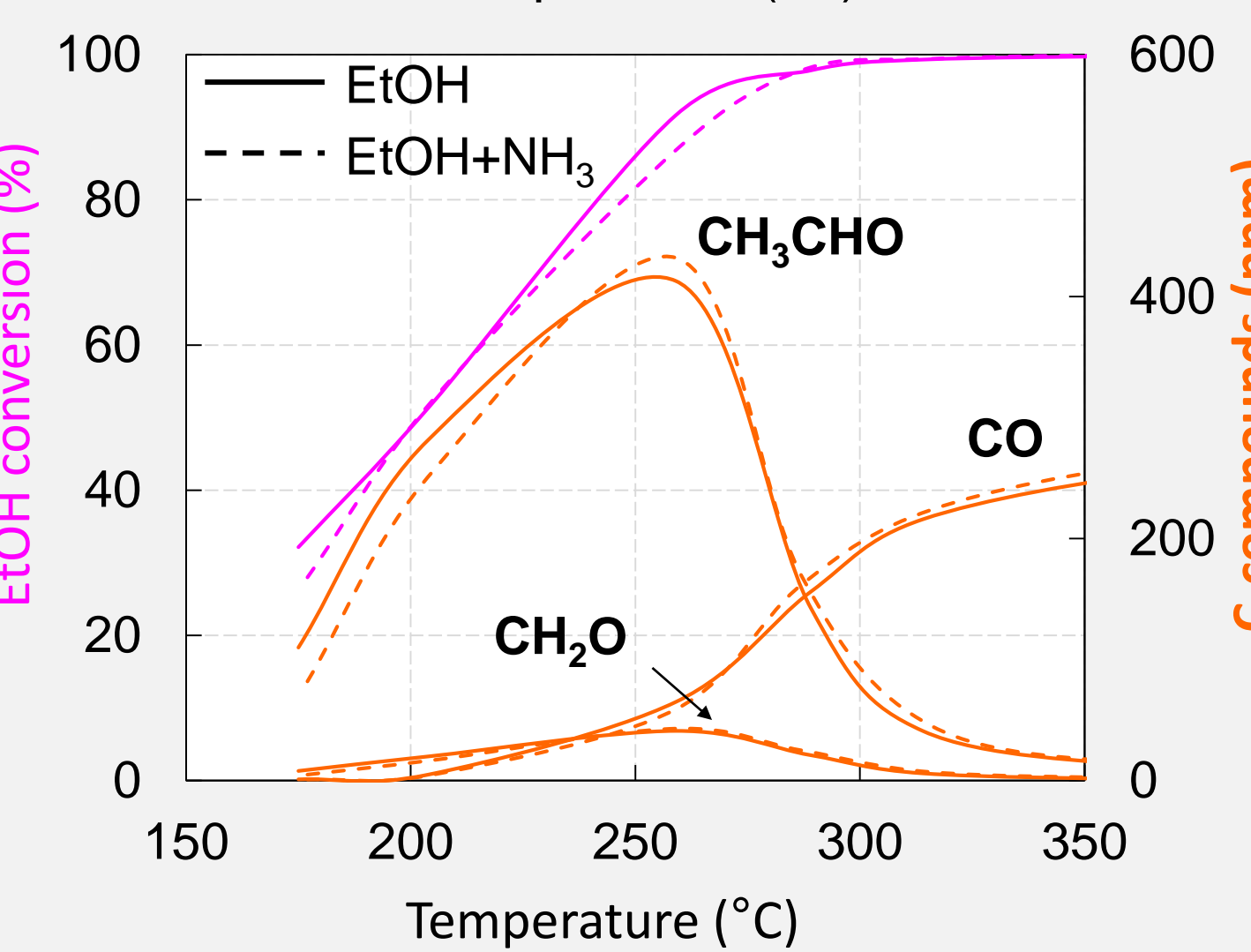
## NH<sub>3</sub> co-feeding on EtOH-SCR on Ag/Al<sub>2</sub>O<sub>3</sub>



### NH<sub>3</sub> co-feeding:

- Remarkable enhancement of the NO<sub>x</sub> conversion at low T

- No effect on ethanol conversion and C-compounds distribution (mainly CH<sub>3</sub>CHO)

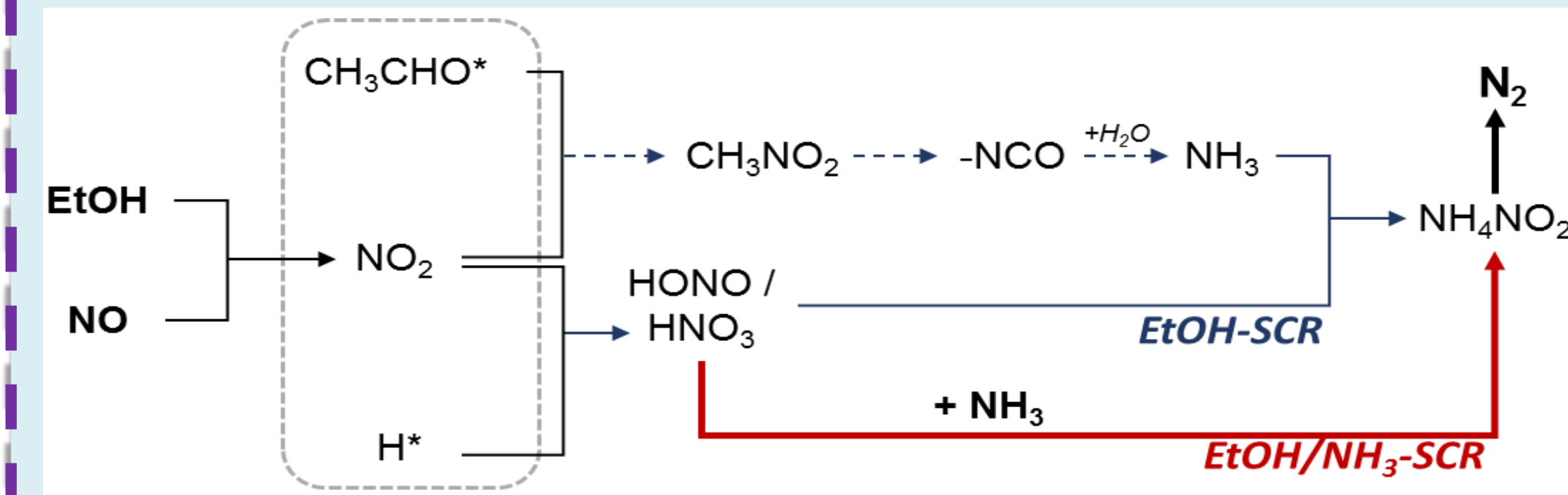


➢ ≈ no NH SCR activity over Ag/Al<sub>2</sub>O<sub>3</sub>

➢ Same improvement if H<sub>2</sub> replaced ethanol (with H<sub>2</sub> amount corresponding to ethanol dehydrogenation to acetaldehyde)

➢ IR *in situ* (200°C): adsorption of (NO-O<sub>2</sub>) after (EtOH-O<sub>2</sub>) preadsorption: ⇒ HNO<sub>x</sub> species (reactive species toward NH<sub>3</sub>)

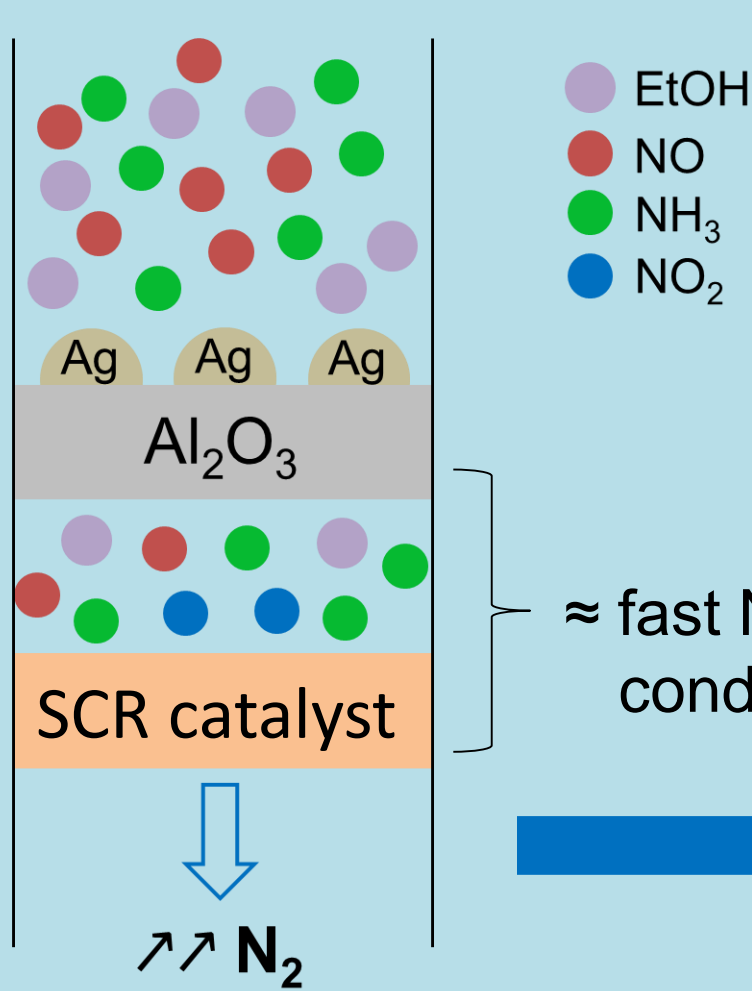
## Reaction pathway in (EtOH+NH<sub>3</sub>)-SCR over Ag/Al<sub>2</sub>O<sub>3</sub>



Barreau M, Tarot M-L, Duprez D, Courtois X, Can F (2018), Appl. Catal. B 220:19-30

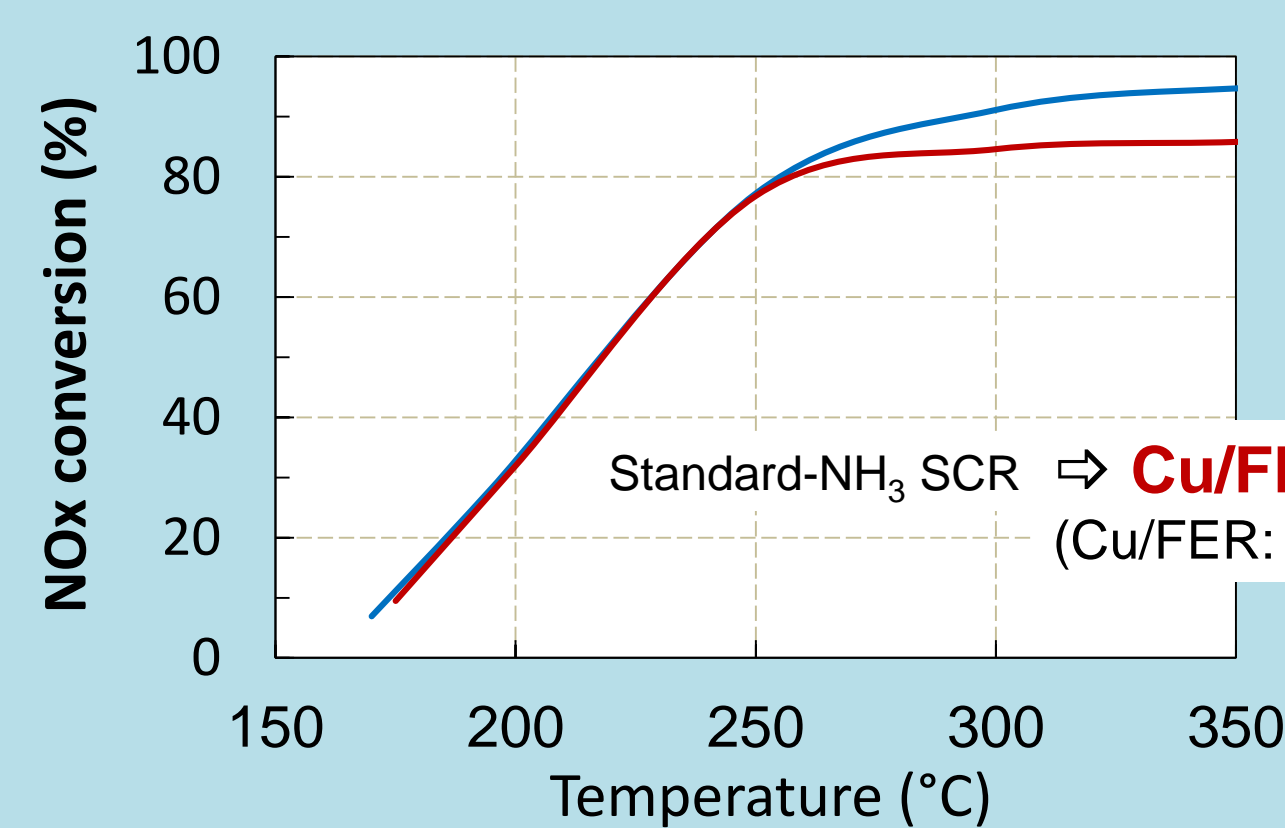
But NO, NO<sub>2</sub> and NH<sub>3</sub> still remain at low T  
⇒ addition of a SCR catalyst

## NO<sub>x</sub> conversion enhancement using a dual-bed configuration: Ag/Al<sub>2</sub>O<sub>3</sub> + (WO<sub>3</sub>/Ce-Zr-O<sub>2</sub> or Cu/FER)

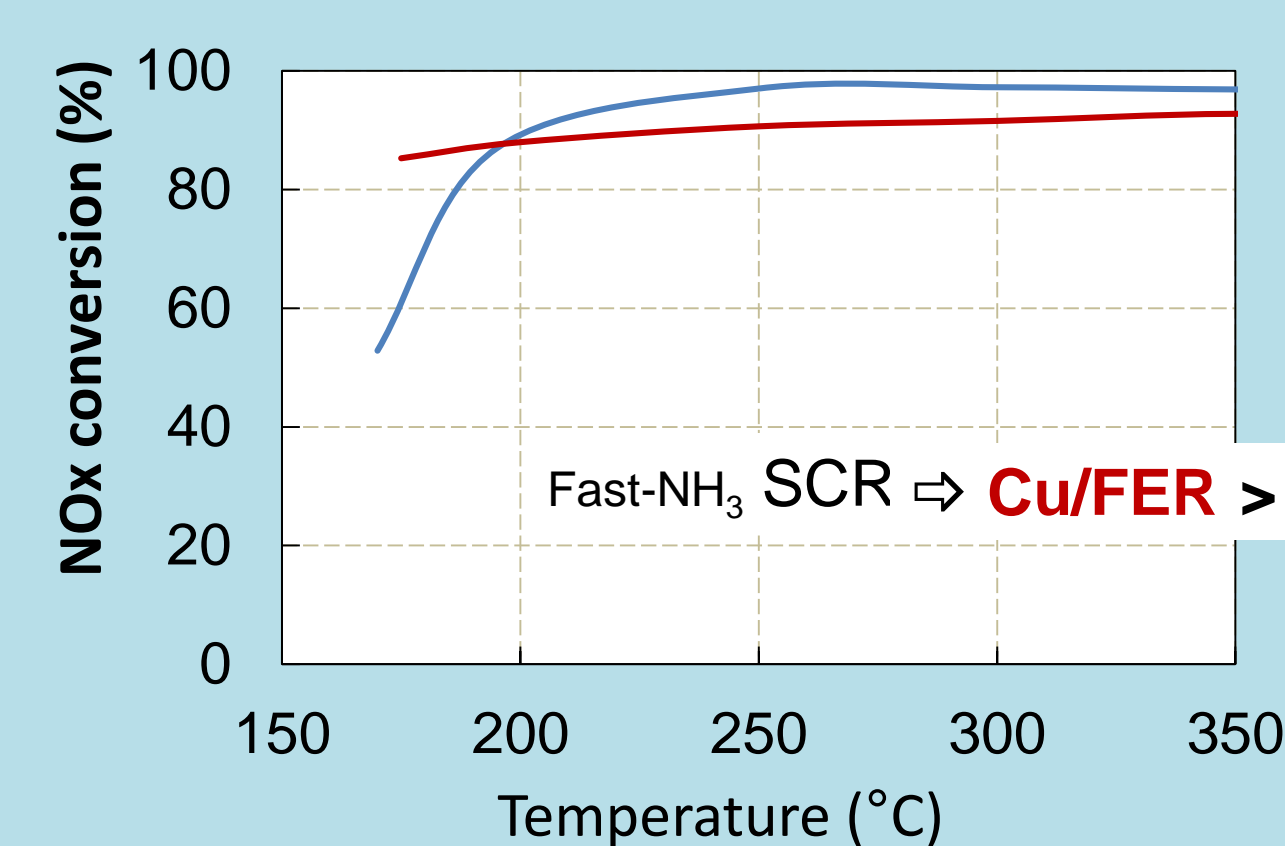


≈ fast NH<sub>3</sub>-SCR condition

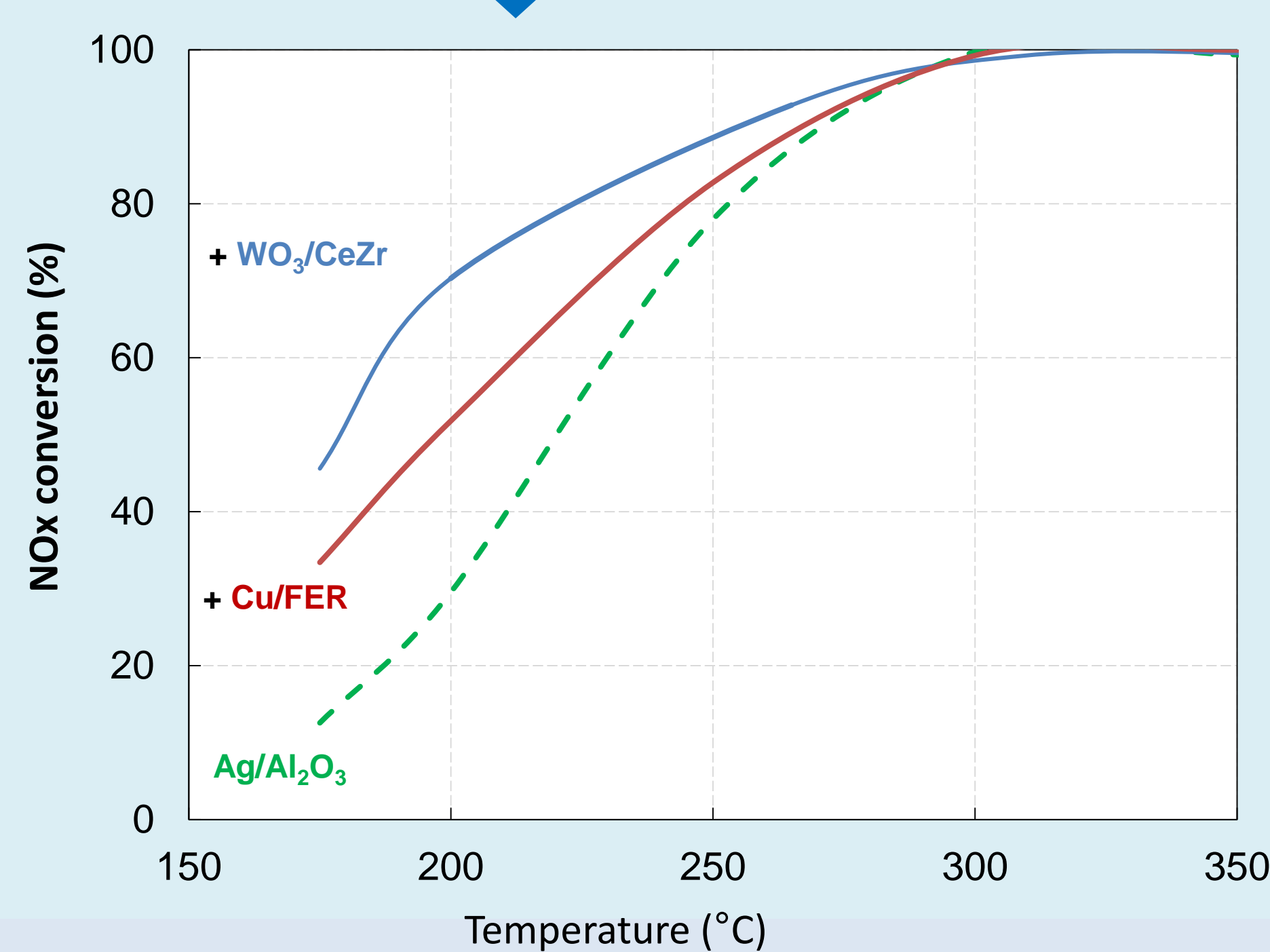
### WO<sub>3</sub>/Ce-Zr-O<sub>2</sub> vs Cu/FER in NH<sub>3</sub> SCR



Standard-NH<sub>3</sub> SCR ⇒ Cu/FER = WO<sub>3</sub>/Ce-Zr-O<sub>2</sub>  
(Cu/FER: conv. NH<sub>3</sub> > conv. NO<sub>x</sub>)

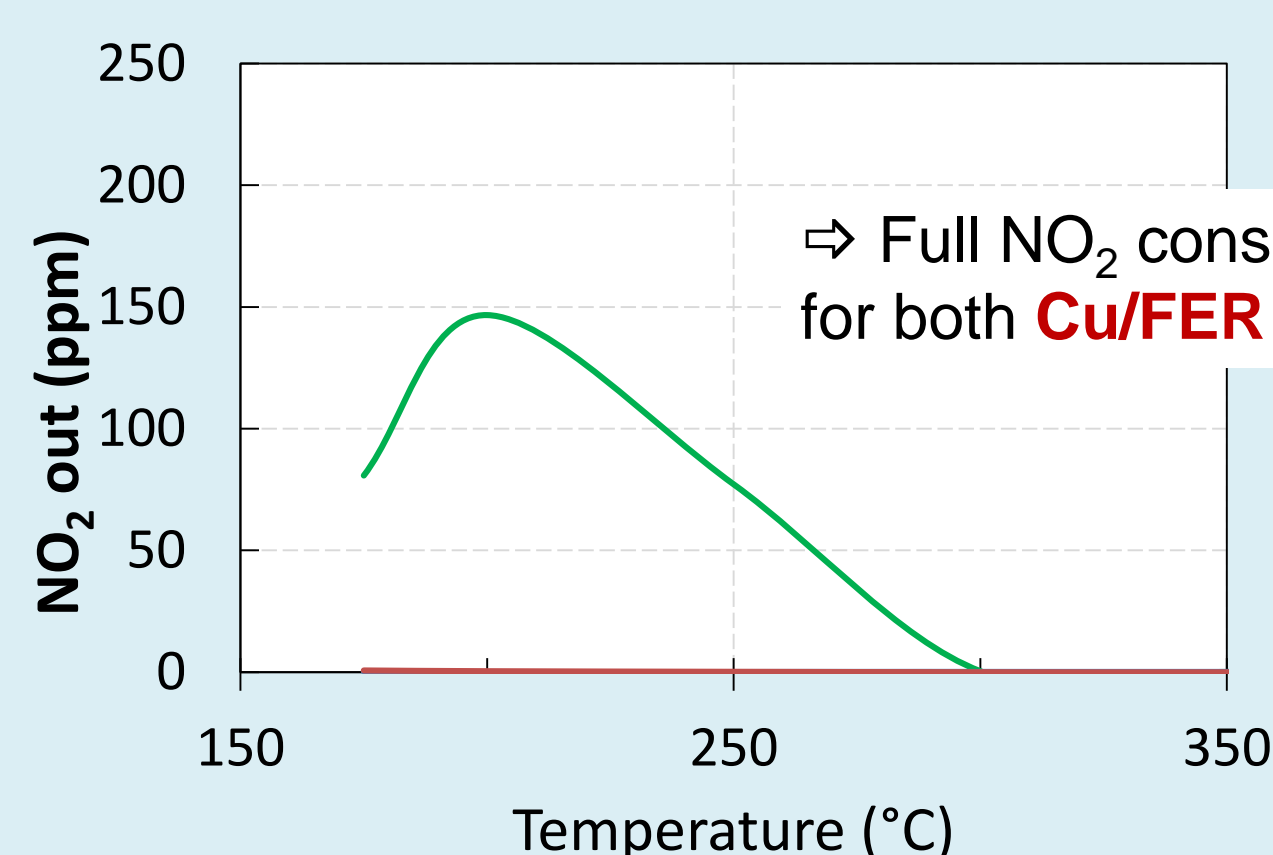


Fast-NH<sub>3</sub> SCR ⇒ Cu/FER > WO<sub>3</sub>/Ce-Zr-O<sub>2</sub> (T < 200°C)

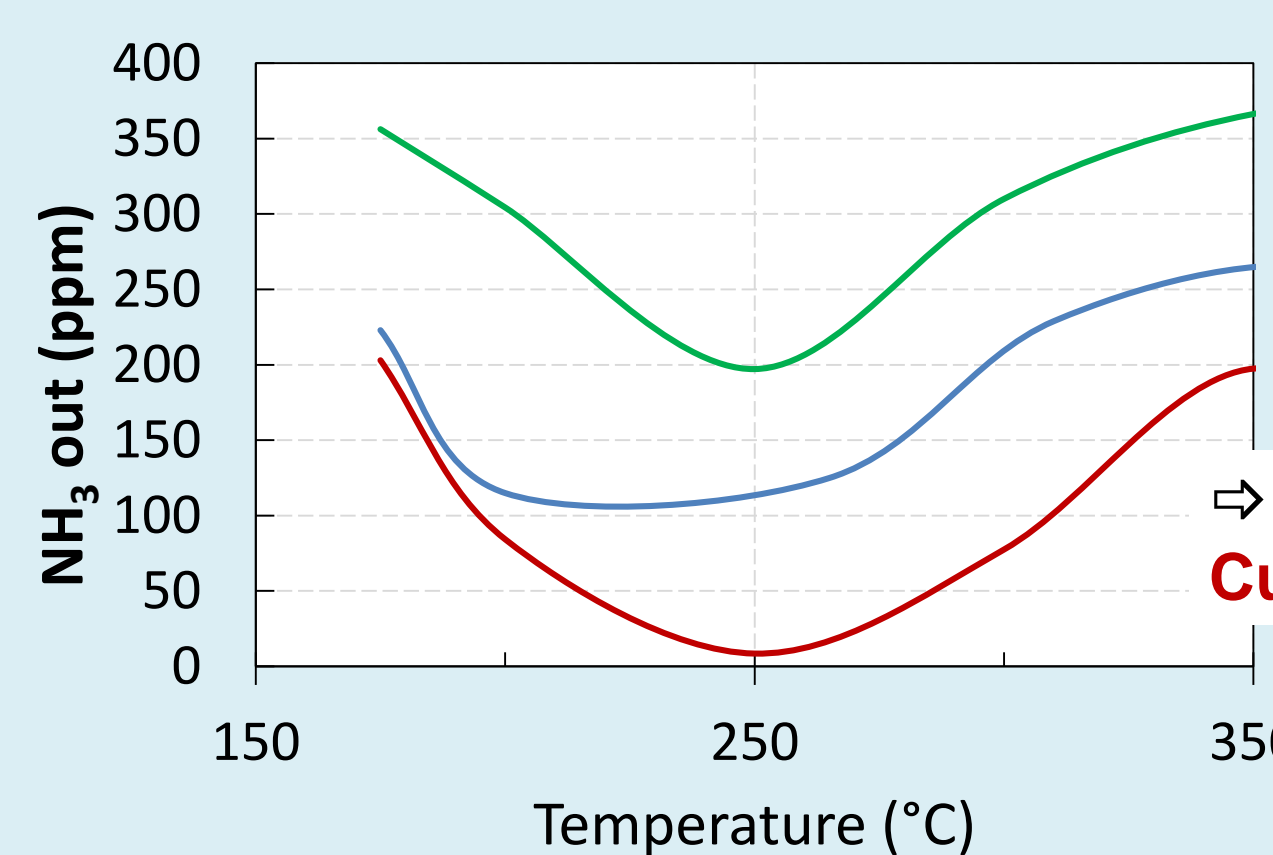


⇒ Remarkable NO<sub>x</sub> conv. at T < 250°C with only NO as NO<sub>x</sub>  
(70% of NO<sub>x</sub> conv. at 200°C while only 9% are achieved in EtOH-SCR)

WO<sub>3</sub>/Ce-Zr-O<sub>2</sub> > Cu/FER

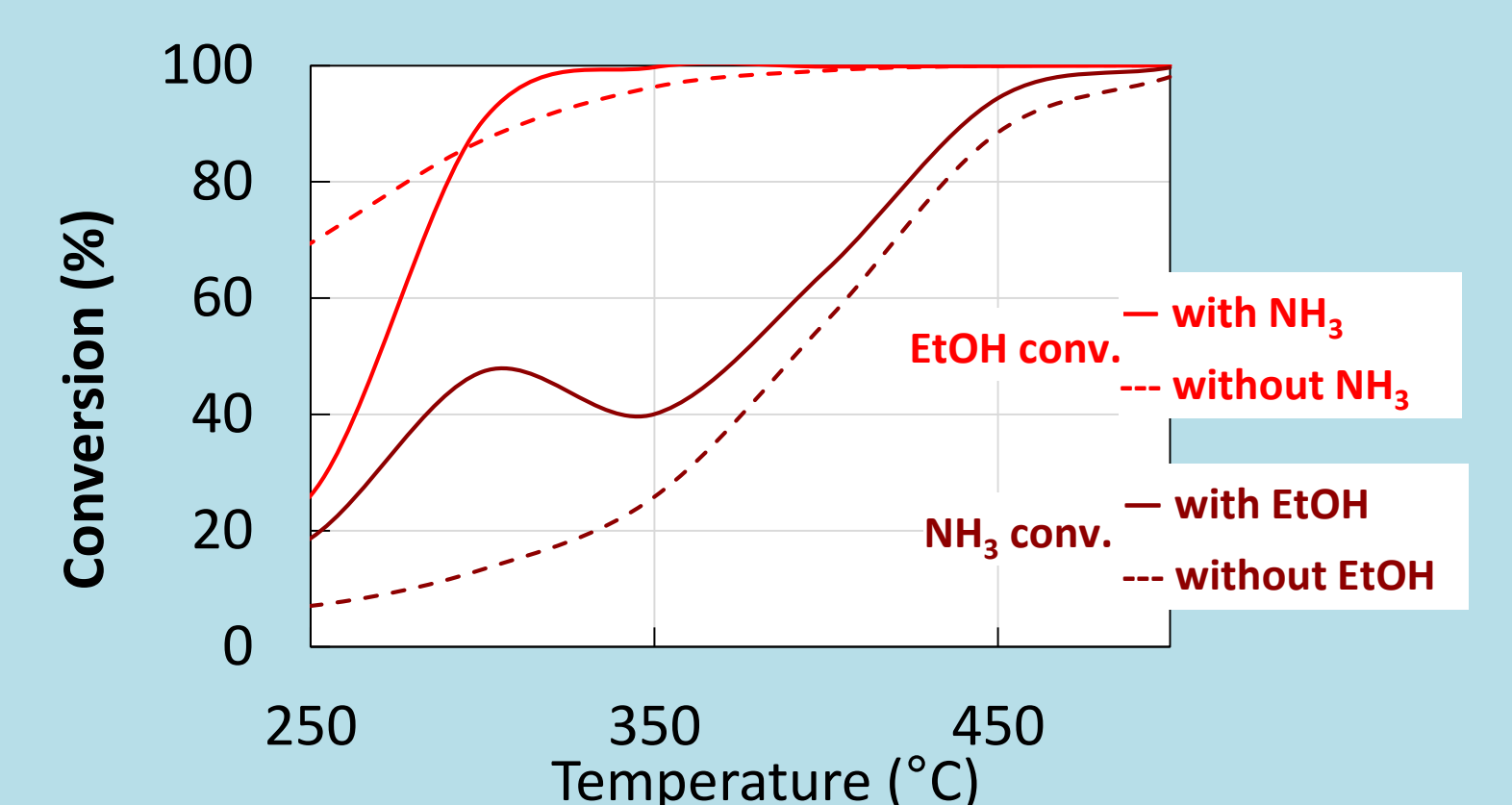


⇒ Full NO<sub>2</sub> consumption from Ag/Al<sub>2</sub>O<sub>3</sub> for both Cu/FER and WO<sub>3</sub>/Ce-Zr-O<sub>2</sub>



⇒ NH<sub>3</sub> consumption: Cu/FER > WO<sub>3</sub>/Ce-Zr-O<sub>2</sub>

### EtOH - NH<sub>3</sub> oxidation tests on Cu/FER



- EtOH conv. ↓ at low T when EtOH was mixed with NH<sub>3</sub>
- NH<sub>3</sub> conv. ↑ when NH<sub>3</sub> was mixed with EtOH

## CONCLUSION

Cu/FER in the dual-bed system led to a lower deNO<sub>x</sub> efficiency than WO<sub>3</sub>/CeZrO<sub>2</sub> despite a promising higher fast-SCR activity at low temperature (vs WO<sub>3</sub>/CeZr), because ammonia and ethanol strongly interacted together on Cu/FER