

Development of a Pilot Scale Reactor for the Selective Oxidation of Ammonia to N₂ and H₂O

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TDA
Research

Overview

Water purification with the VPCAR.

- Advantages and challenges.

Objectives of Phase II Project.

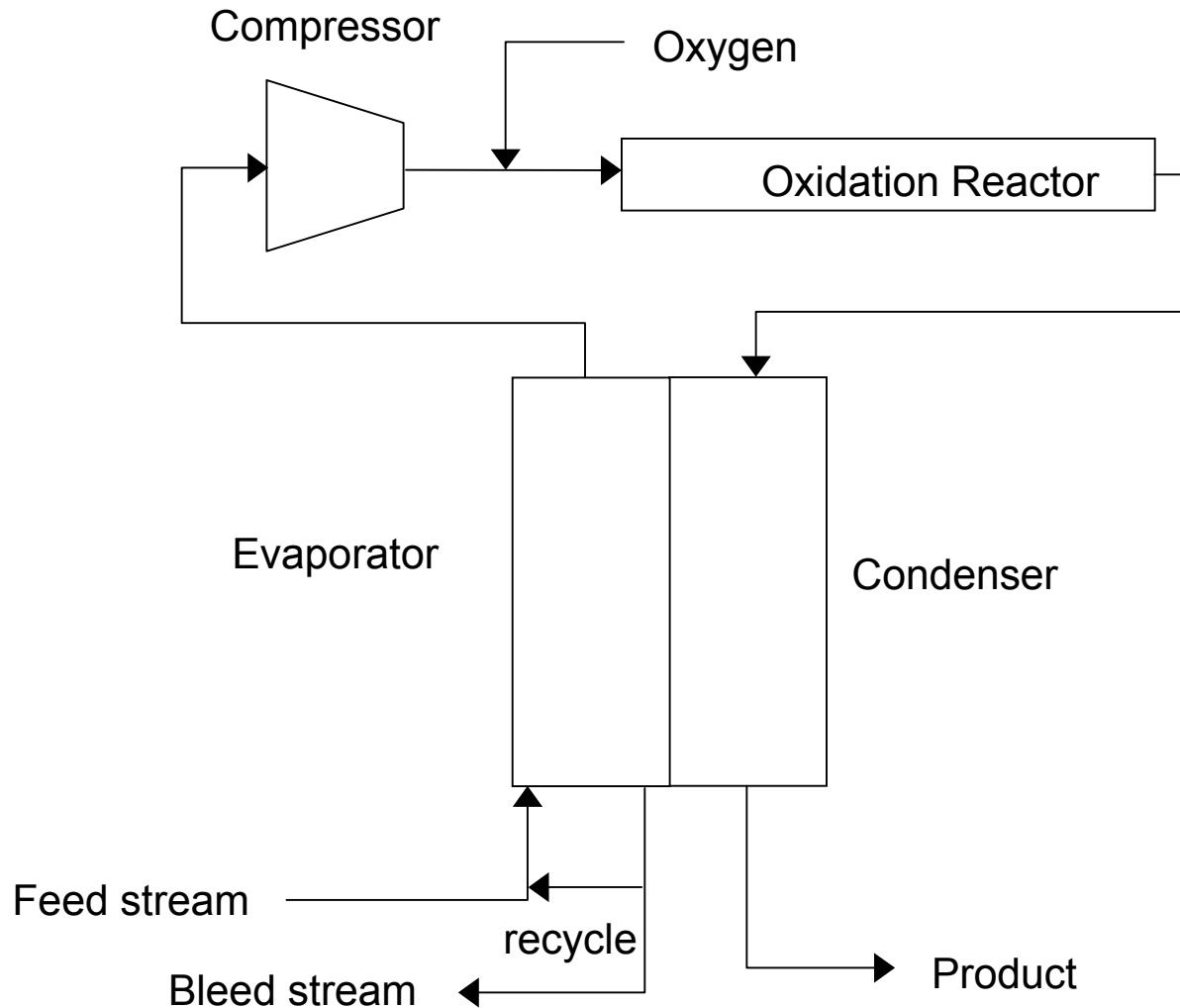
Experimental results to date.

- Catalyst screening for NH_3 oxidation activity.
- NO_x formation.
- Kinetics and rate model.

Design of the pilot scale system.

Hydrocarbon oxidation tests.

Schematic of the VPCAR System



Simplified from Flynn and Borchers, 2000

Advantages and Challenges Associated of the VPCAR

Biggest advantage is that it has no resupply requirements and therefore desirable on a long term mission.

However, volatile components such as ammonia (SMAC = 7 mg/m³, Perry 1998) must be oxidized in the catalyst bed.

Platinum catalysts are good oxidation catalysts, but they can convert ammonia to NO_x (NO and NO₂, SMAC = 5.0 and 0.94 mg/m³).

In Phase I, TDA prepared and tested a series of platinum catalysts that were active for ammonia oxidation and did not produce NO_x.

- Platinum loadings ranged from 0.25 to 3.0 wt%
- Modifier loadings ranged from 3 to 15 wt%.

Goals of Phase II Research

Task 1. Optimize catalyst formulation.

- NH_3 Oxidation (TDA and HSSSI).
- Optimize catalyst for hydrocarbon oxidation (TDA).

Task 2. Catalyst Kinetics

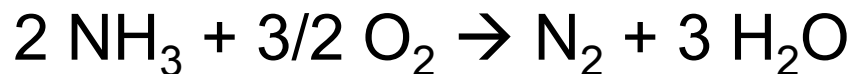
Task 3. Pilot Scale Reactor Design and Testing.

Task 4. Full Scale Reactor Modification (HSSSI).

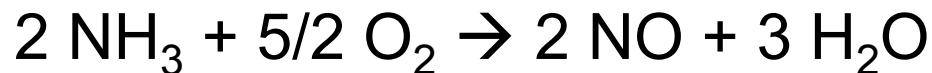
Task 5. Test Full Scale Unit (HSSSI)

Oxidation of NH₃

Selective oxidation of ammonia to nitrogen.

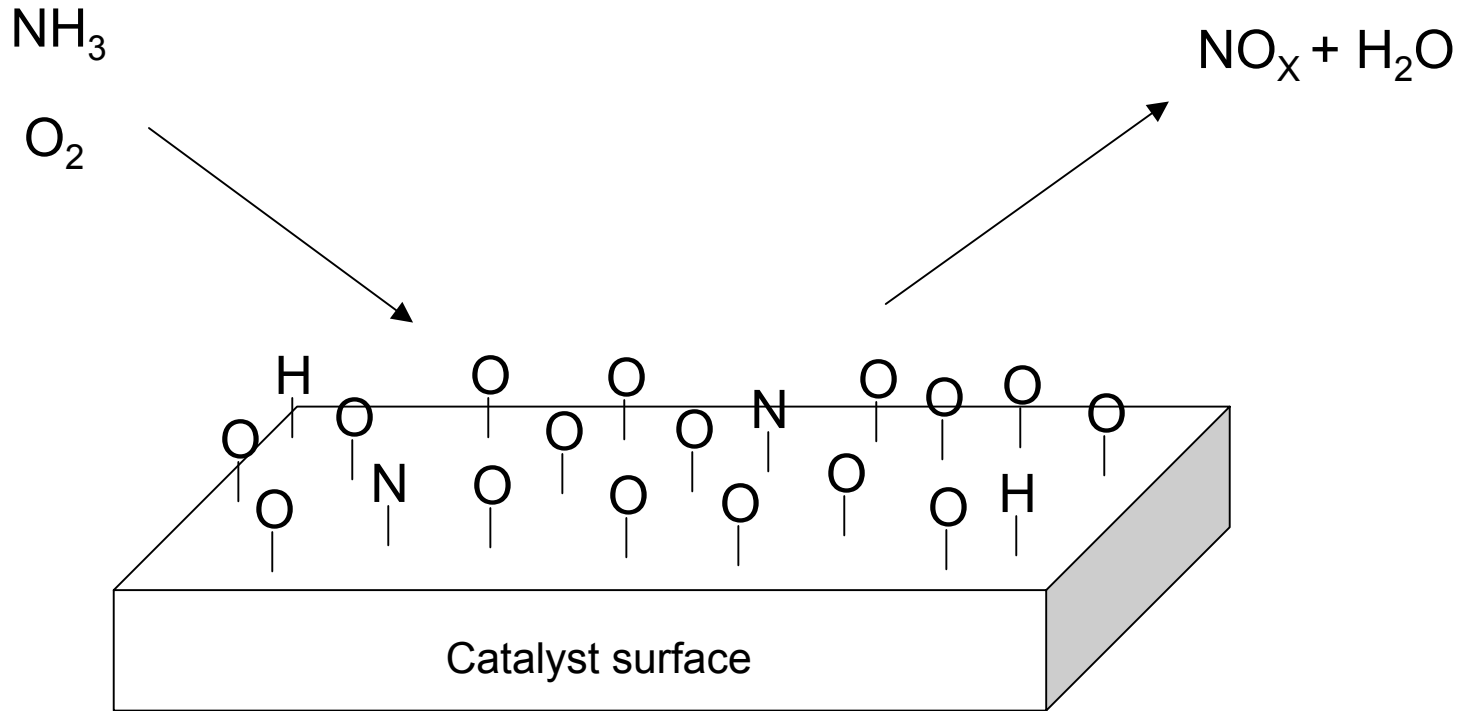


Oxidation of ammonia can also produce NO and NO₂.



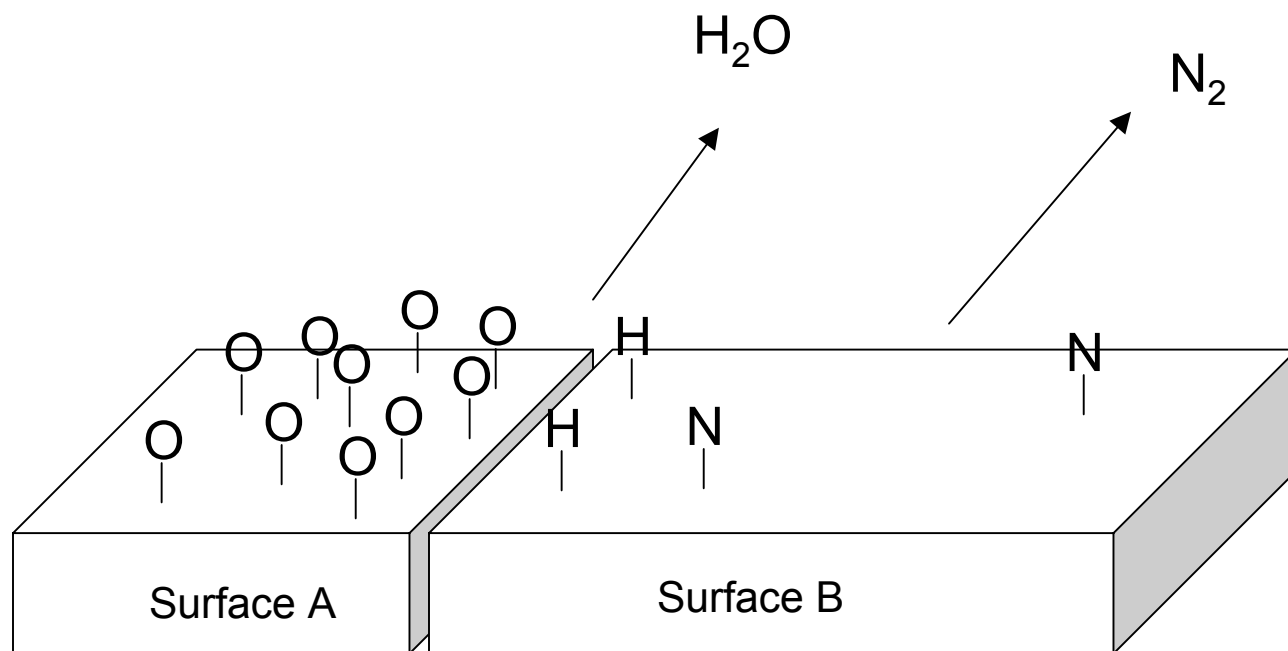
All four reactions can occur; we want to identify catalysts that are selective for N₂.

Why NO_x is Produced During Ammonia Oxidation



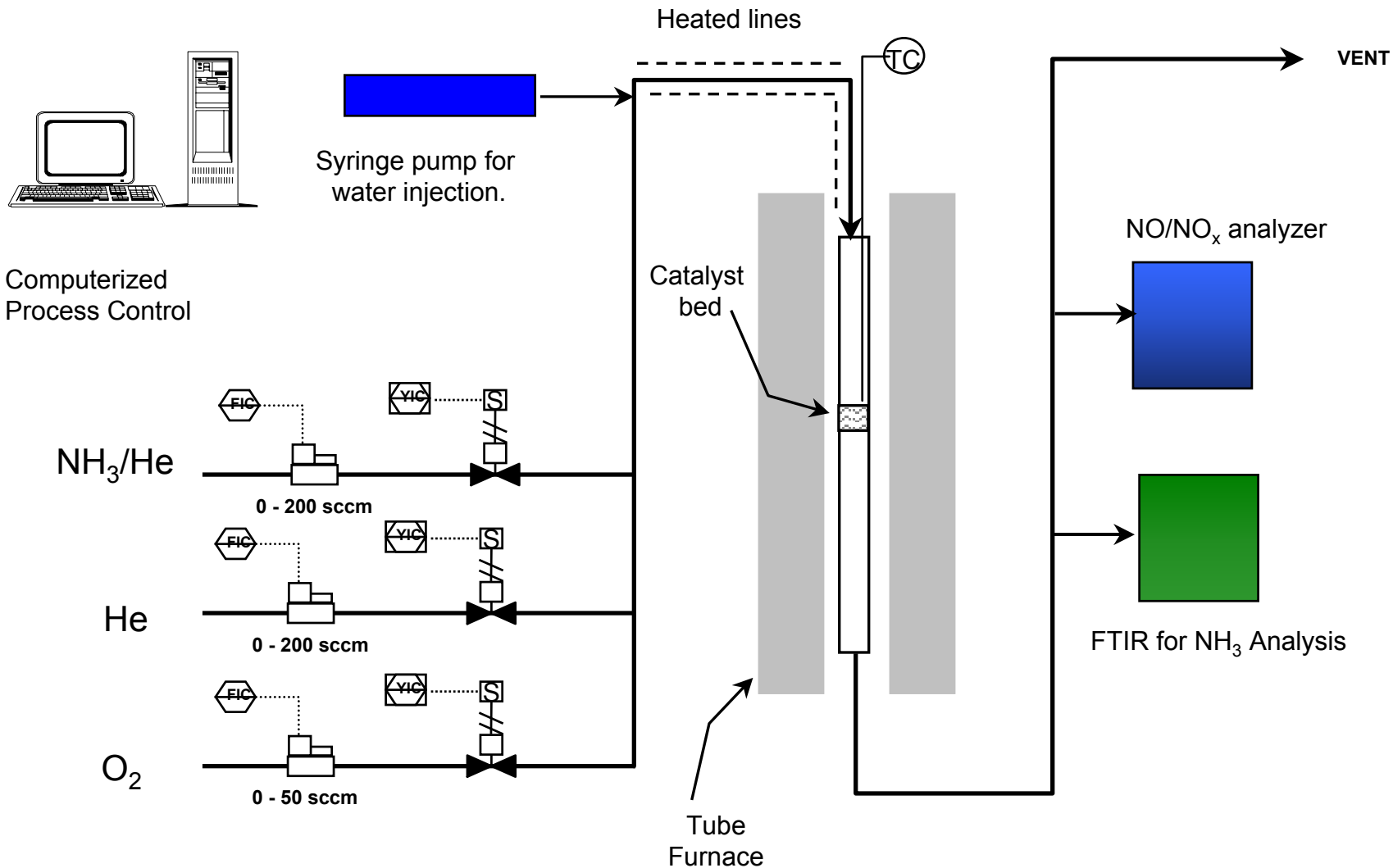
In excess oxygen, there is a high probability that N and O will react to form NO or NO_2 .

Simplified Schematic of Catalyst that is Selective for Nitrogen Production

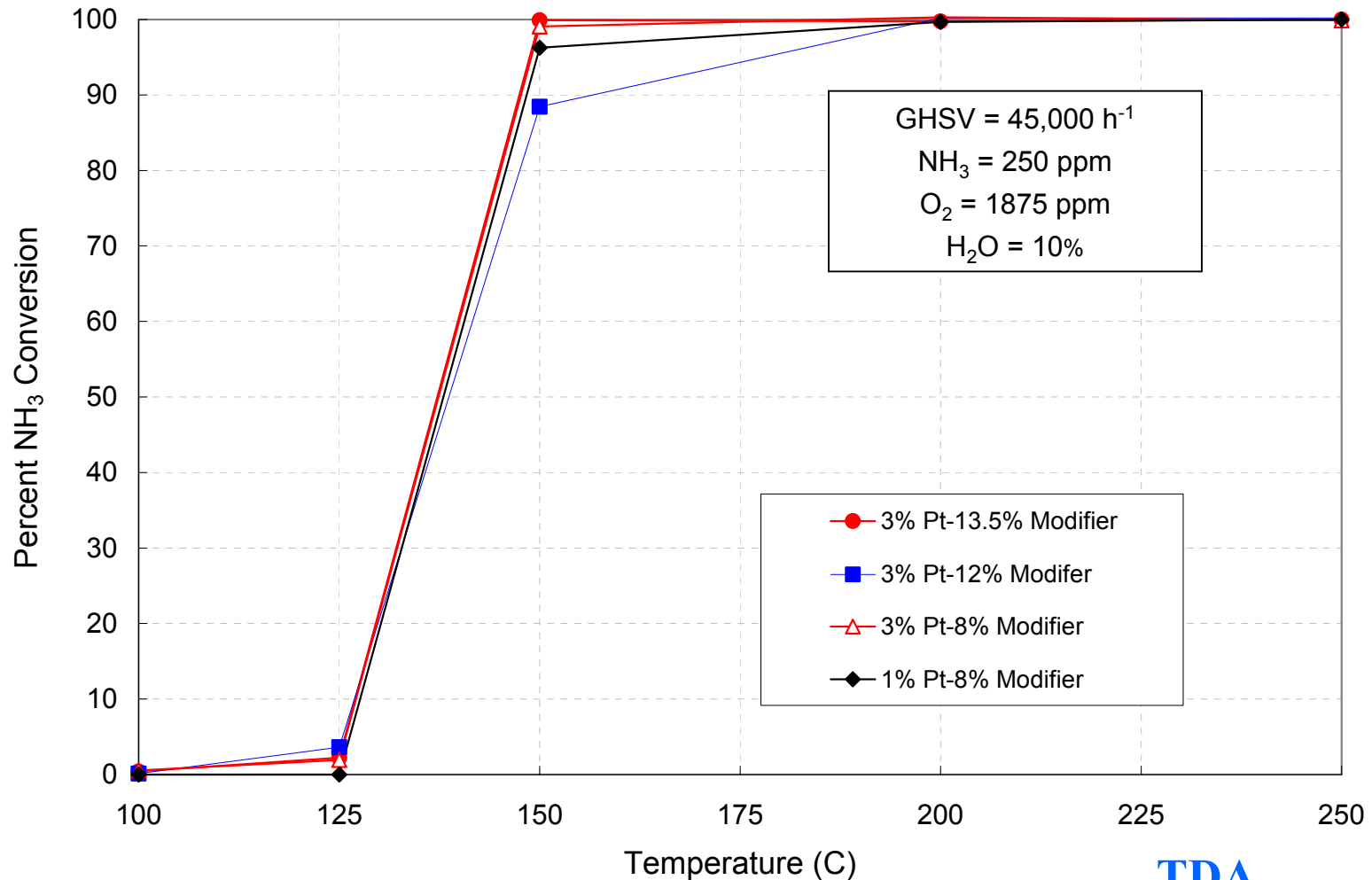


Separate adsorbed O from adsorbed N

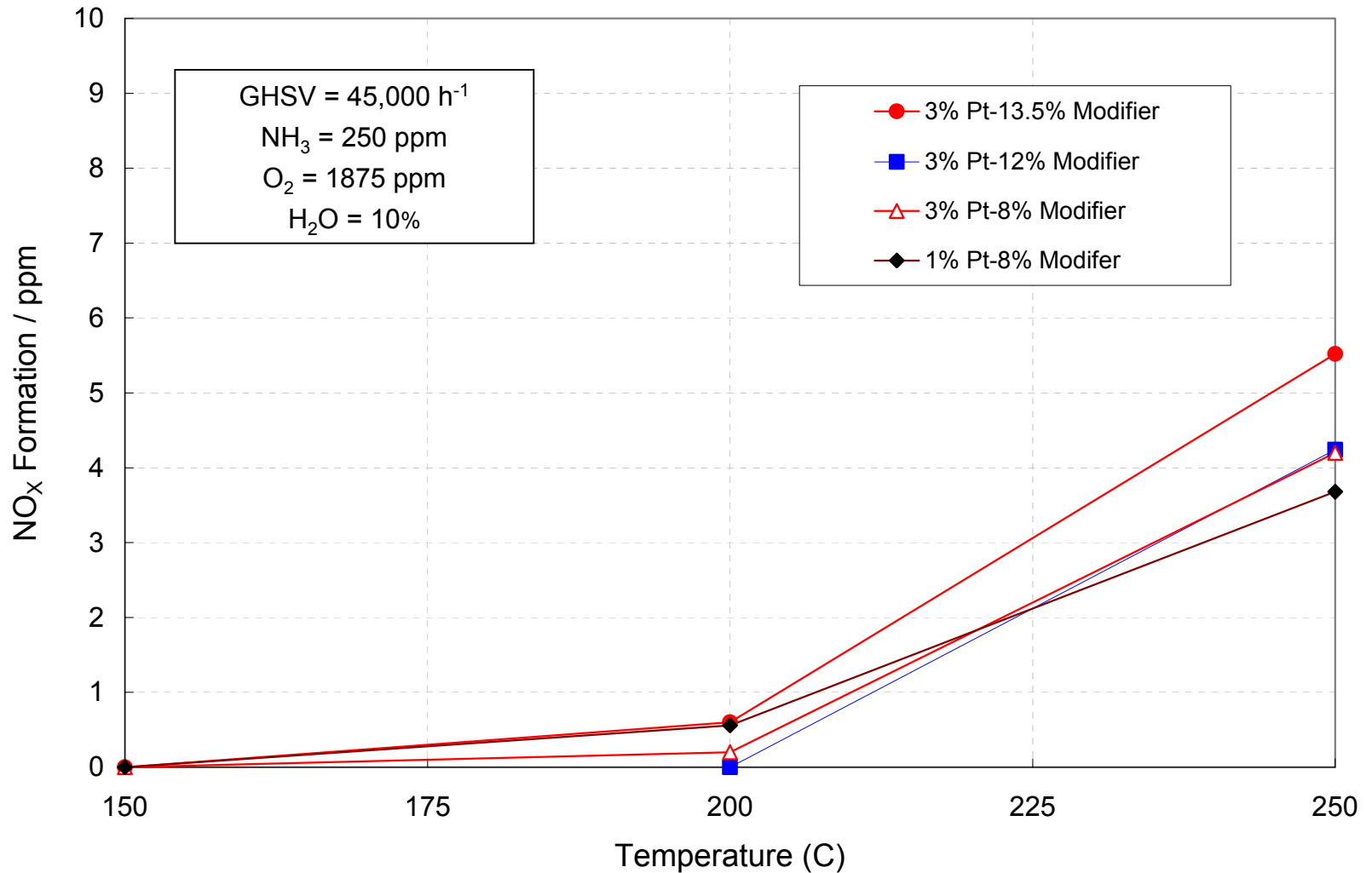
Test Apparatus



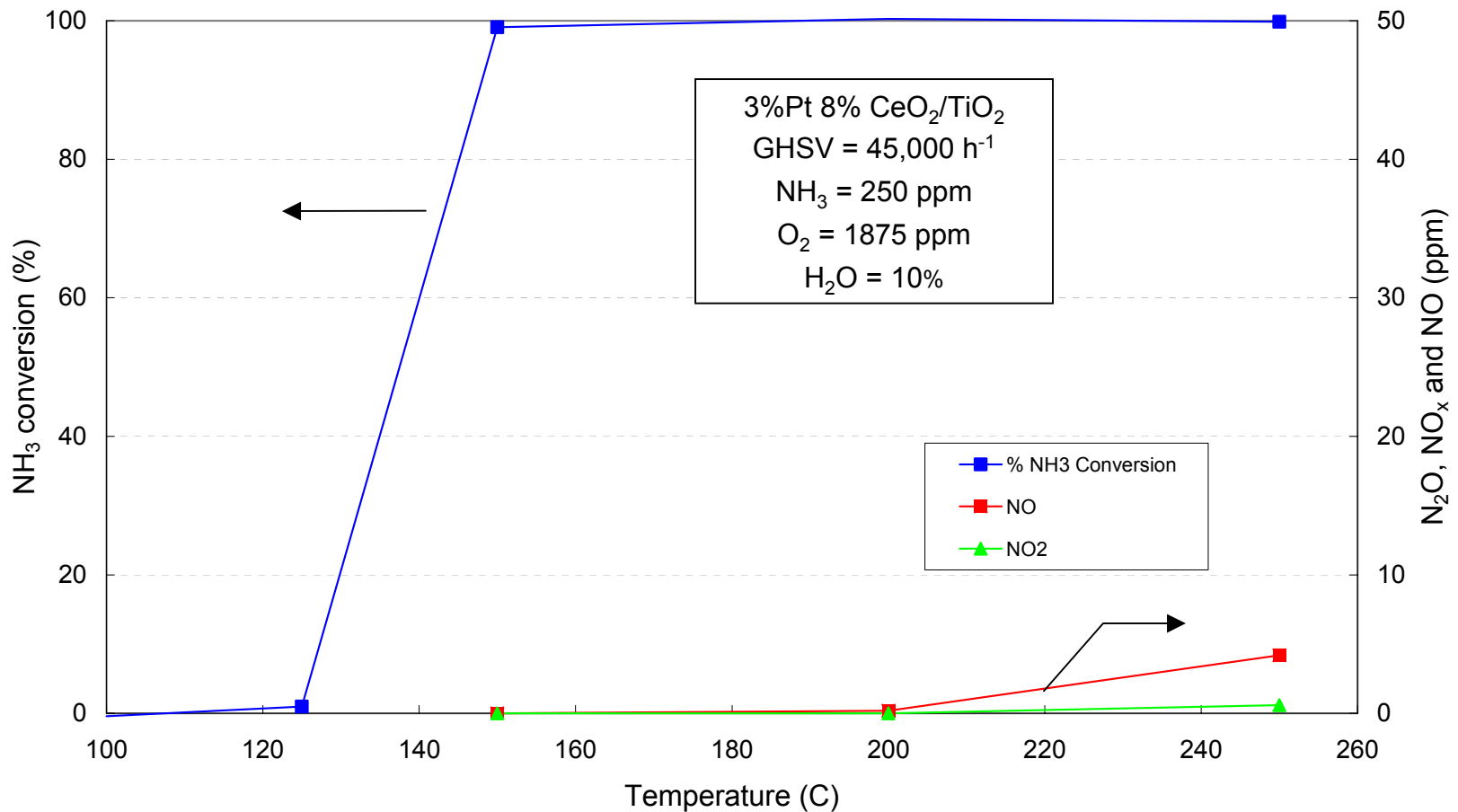
Catalyst Formulations Are Active for Ammonia Oxidation



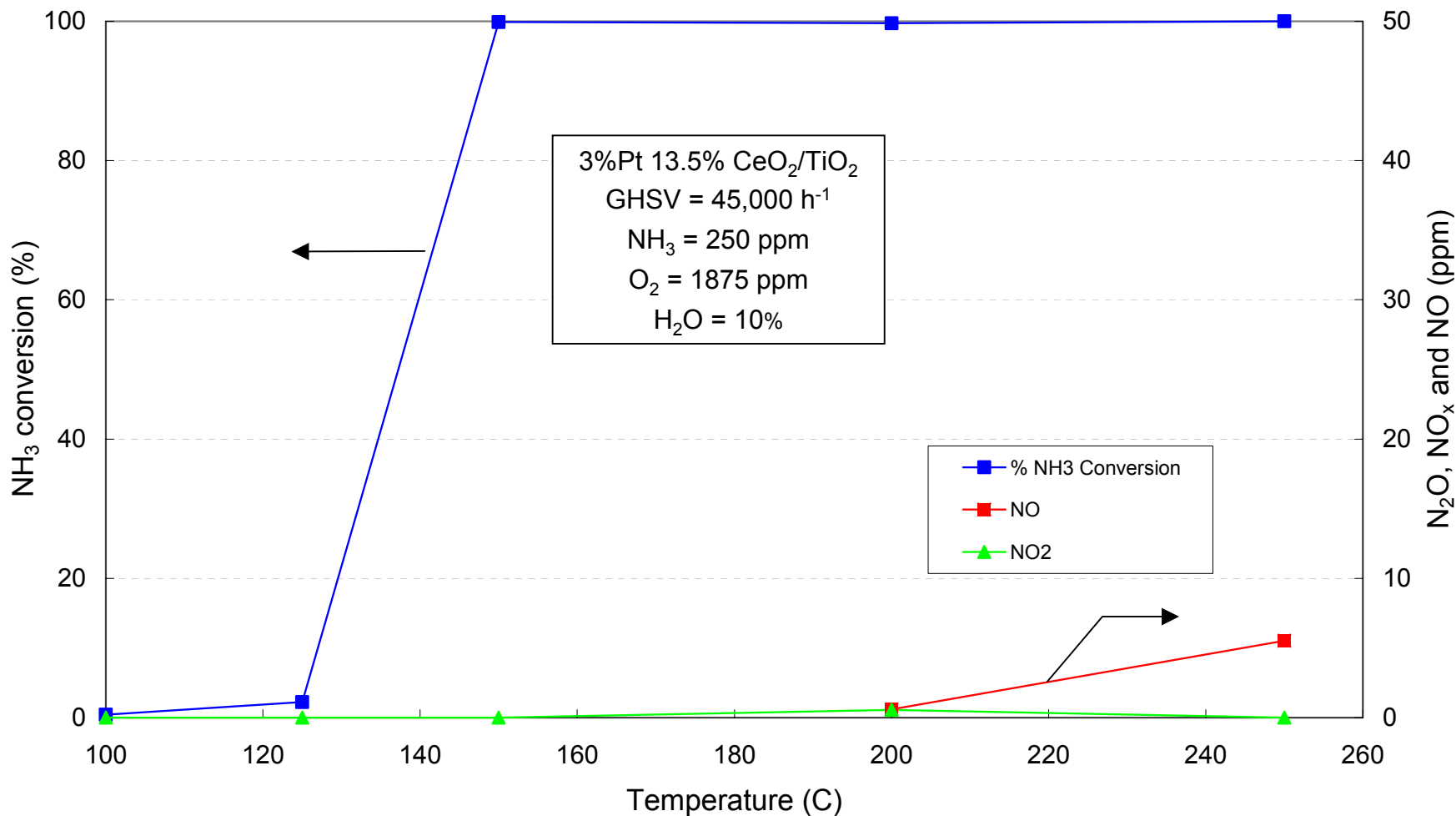
Low NO_x Production up to 200°C



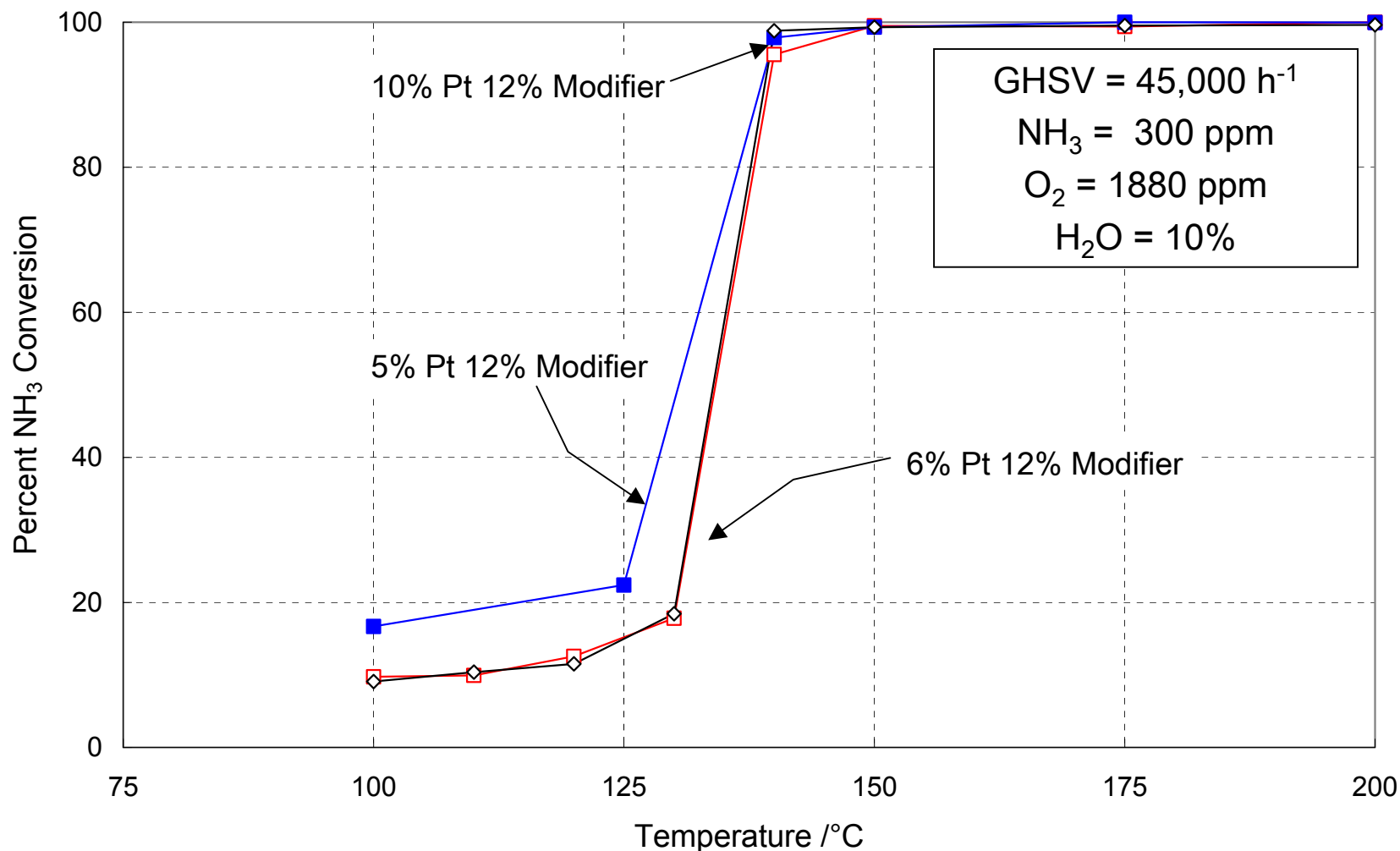
3% Pt 8% Modifier Produces High Ammonia Conversion with Low NO_x



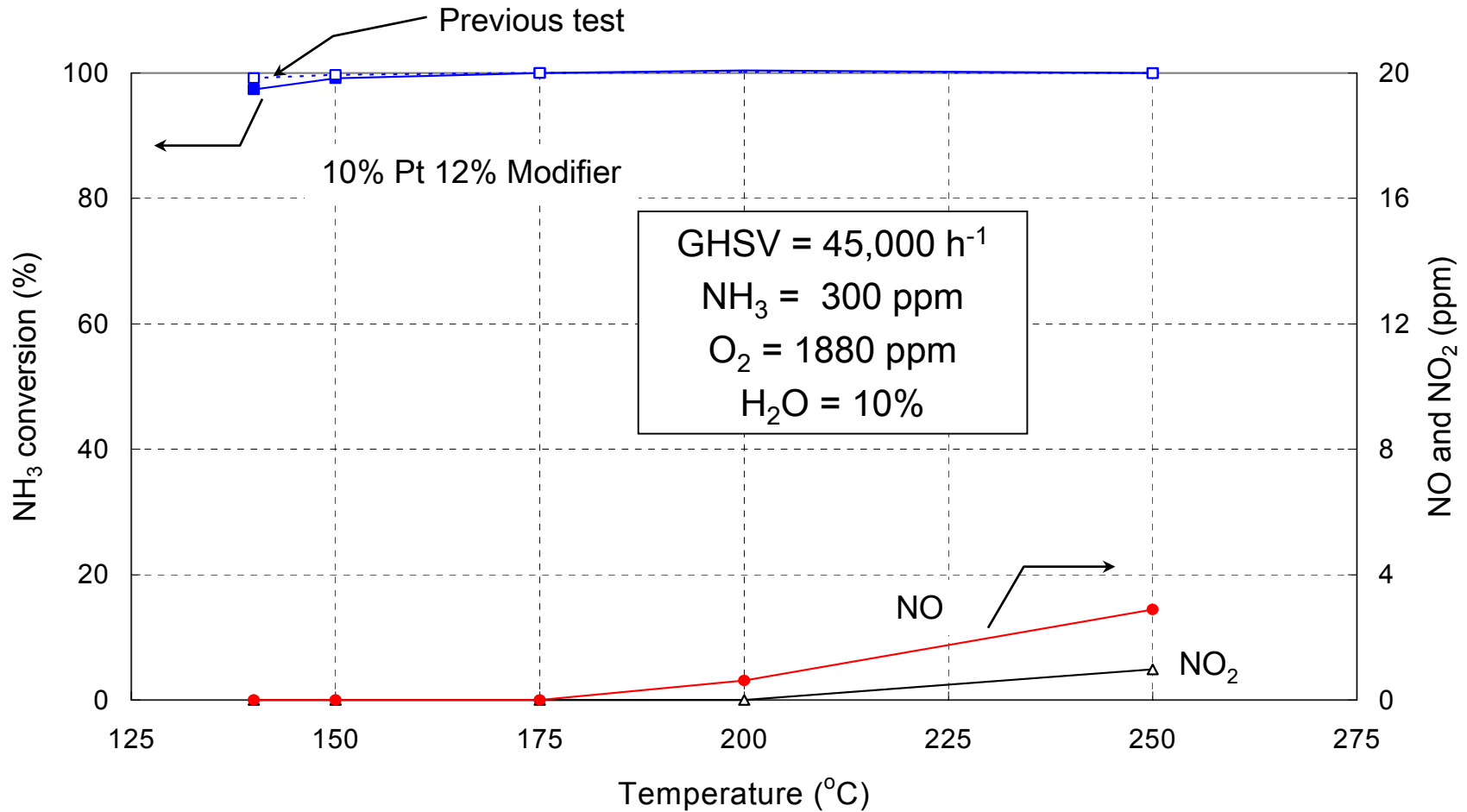
3% Pt 13.5% Modifier is Also an Effective Catalyst



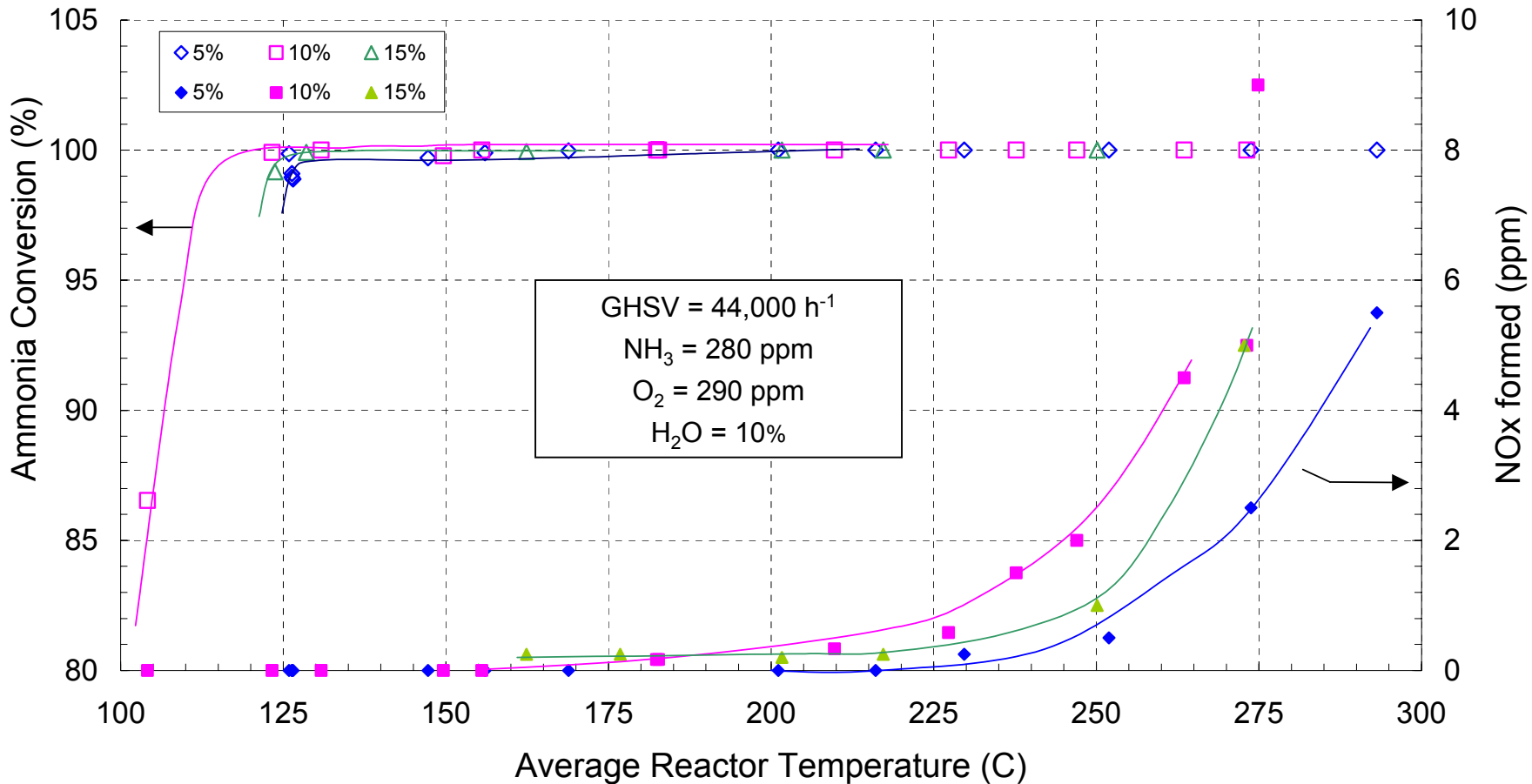
Higher Metal Loadings Produce 100% Conversion at 140°C



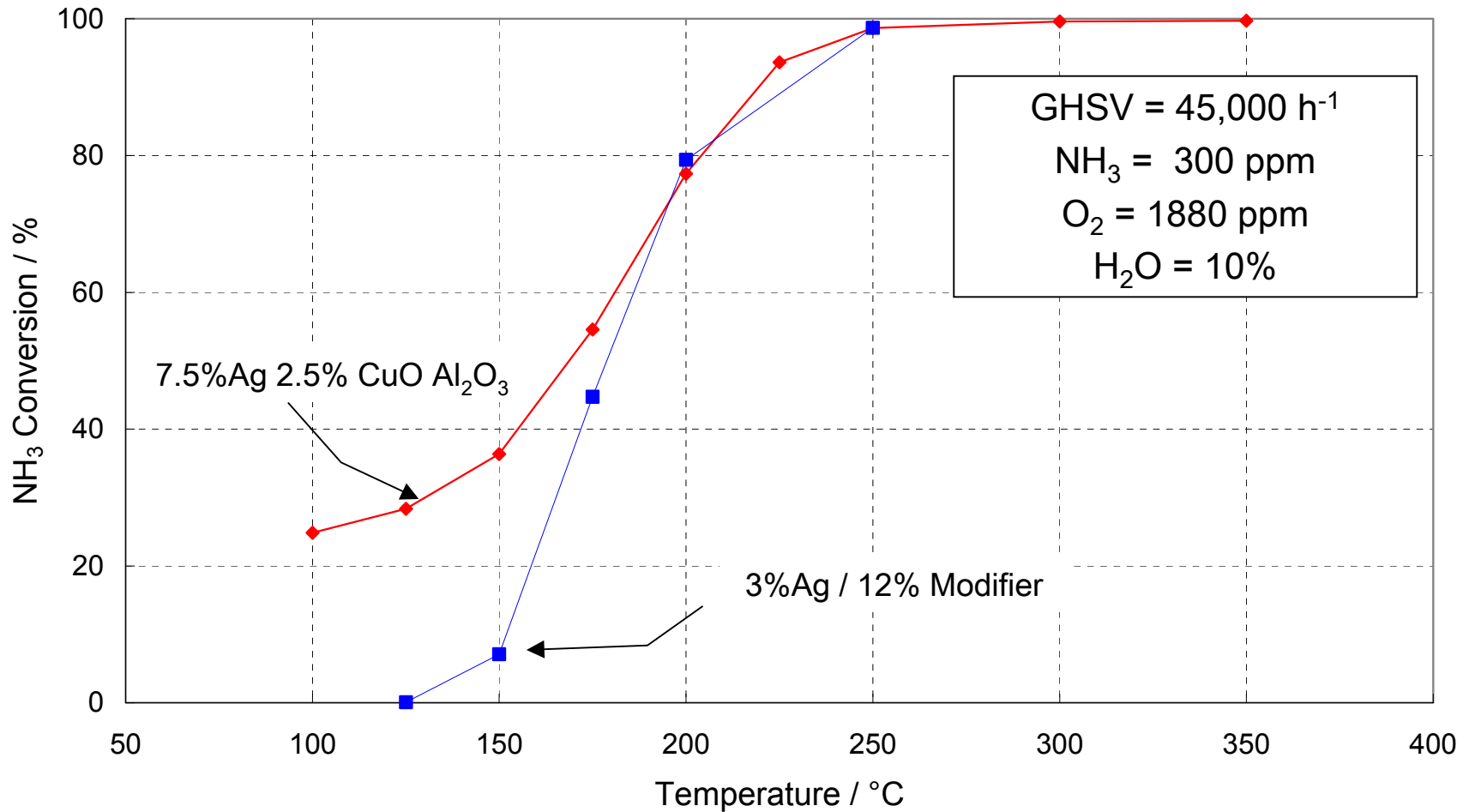
Better Ammonia Conversion without Increasing NO_x



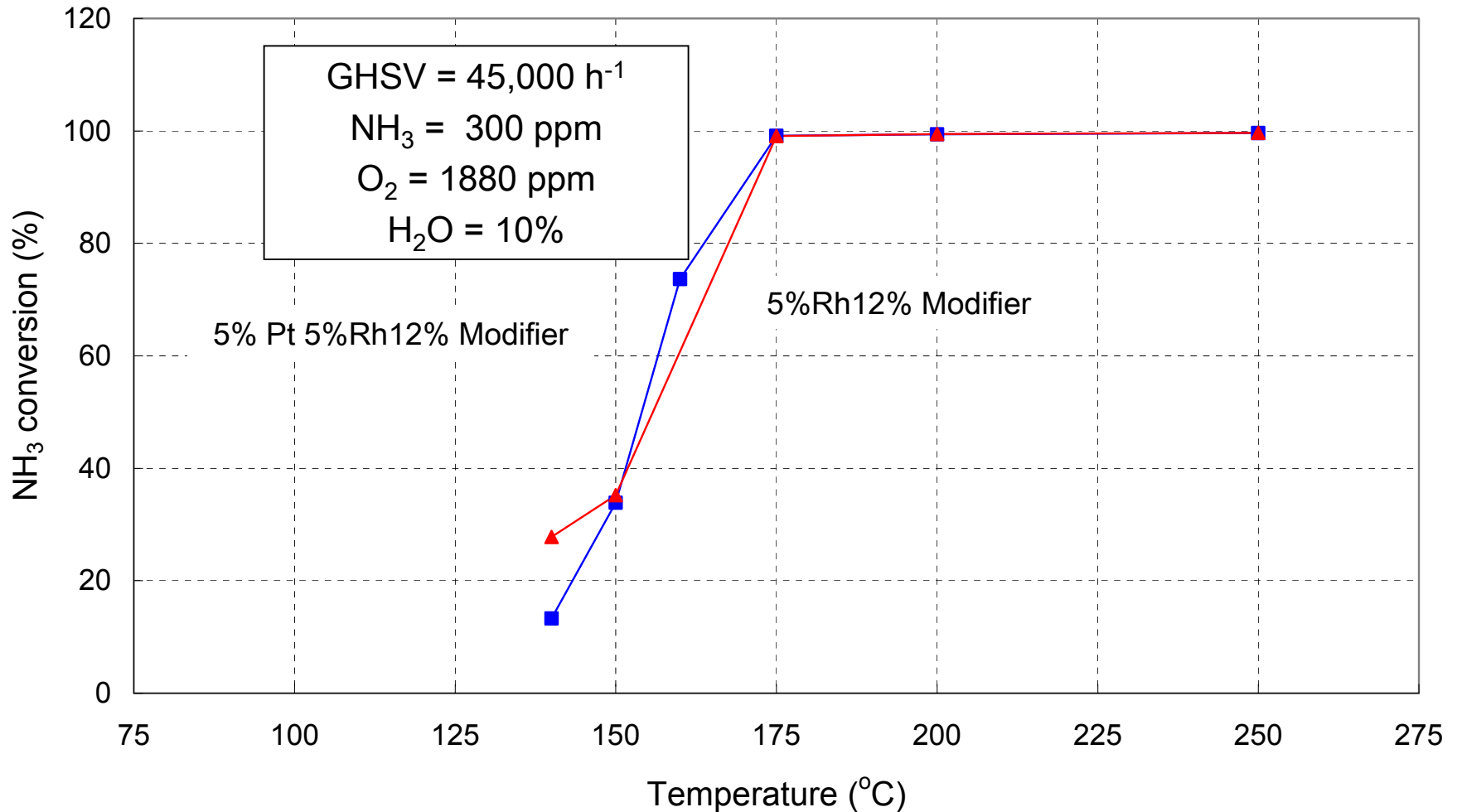
HSSSI Catalysts are Active and Do Not Produce NO_x Below 200°C



Cu and Ag Catalysts have Lower Activity for NH₃ Oxidation



Pt Rh Catalysts are More Active than Ag or Cu Catalysts



Predict Catalyst Performance in the Pilot Scale Reactor

Catalytic reactor is 2-in ID x 8-in long

- (25.1 in³, 0.414 liters, or 0.14 of full scale).

Operating pressure = 92 torr.

Nominal water flow = 1.8 lb/h.

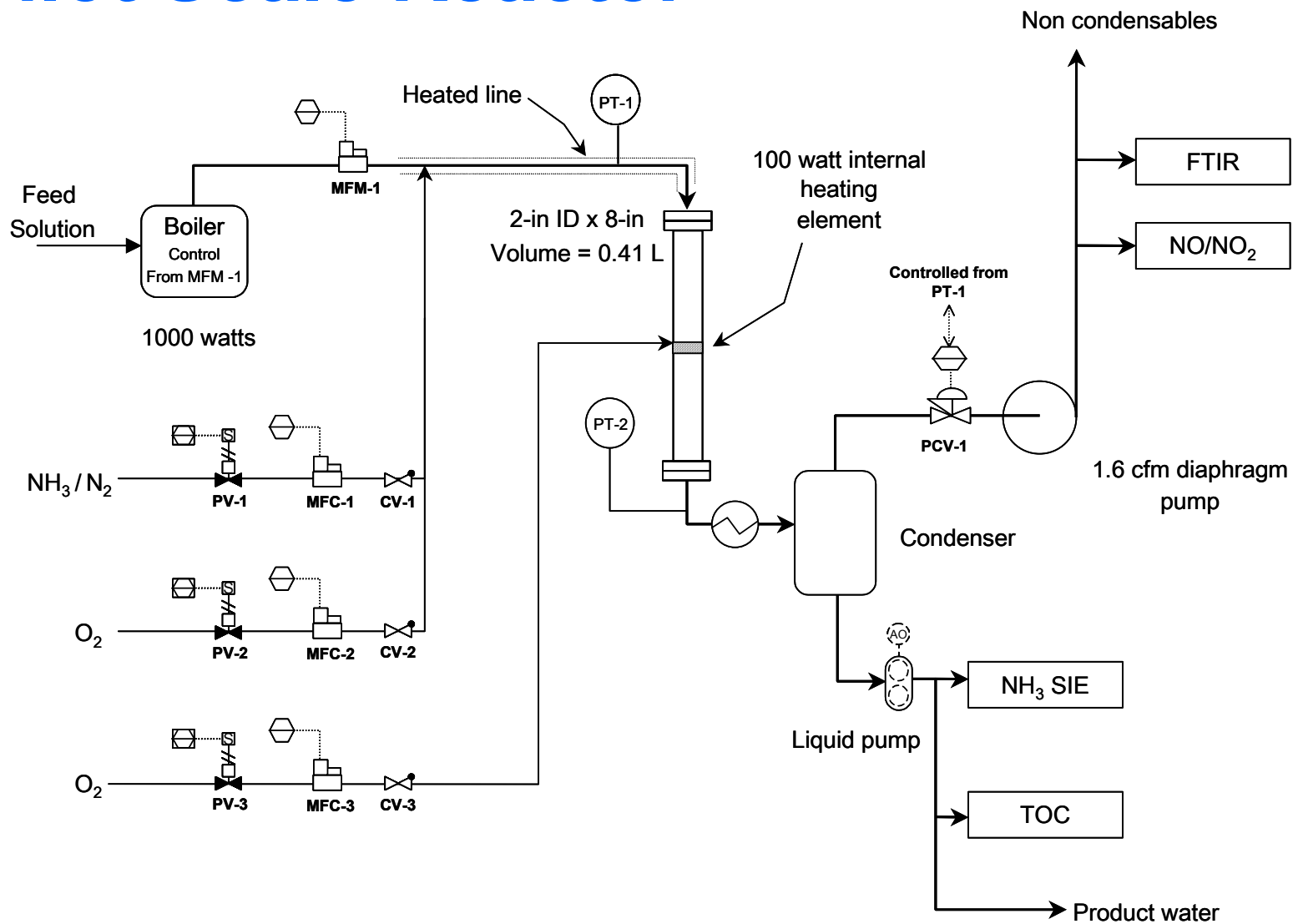
O₂ flow = 0.0182 lb/h (100 sccm).

Operating temperature = 140 - 160°C.

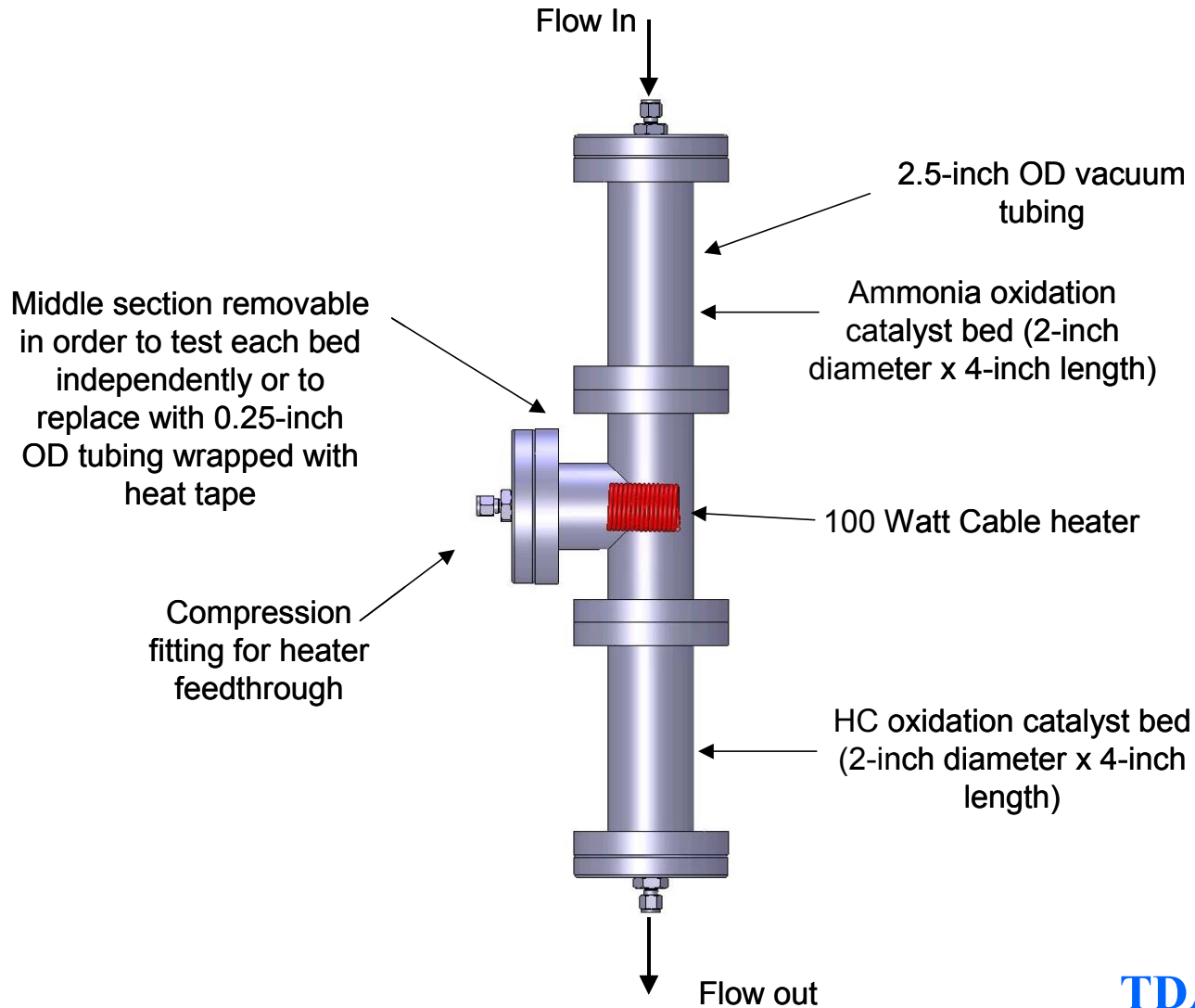
Front of reactor - ammonia oxidation.

Back of reactor - HC oxidation.

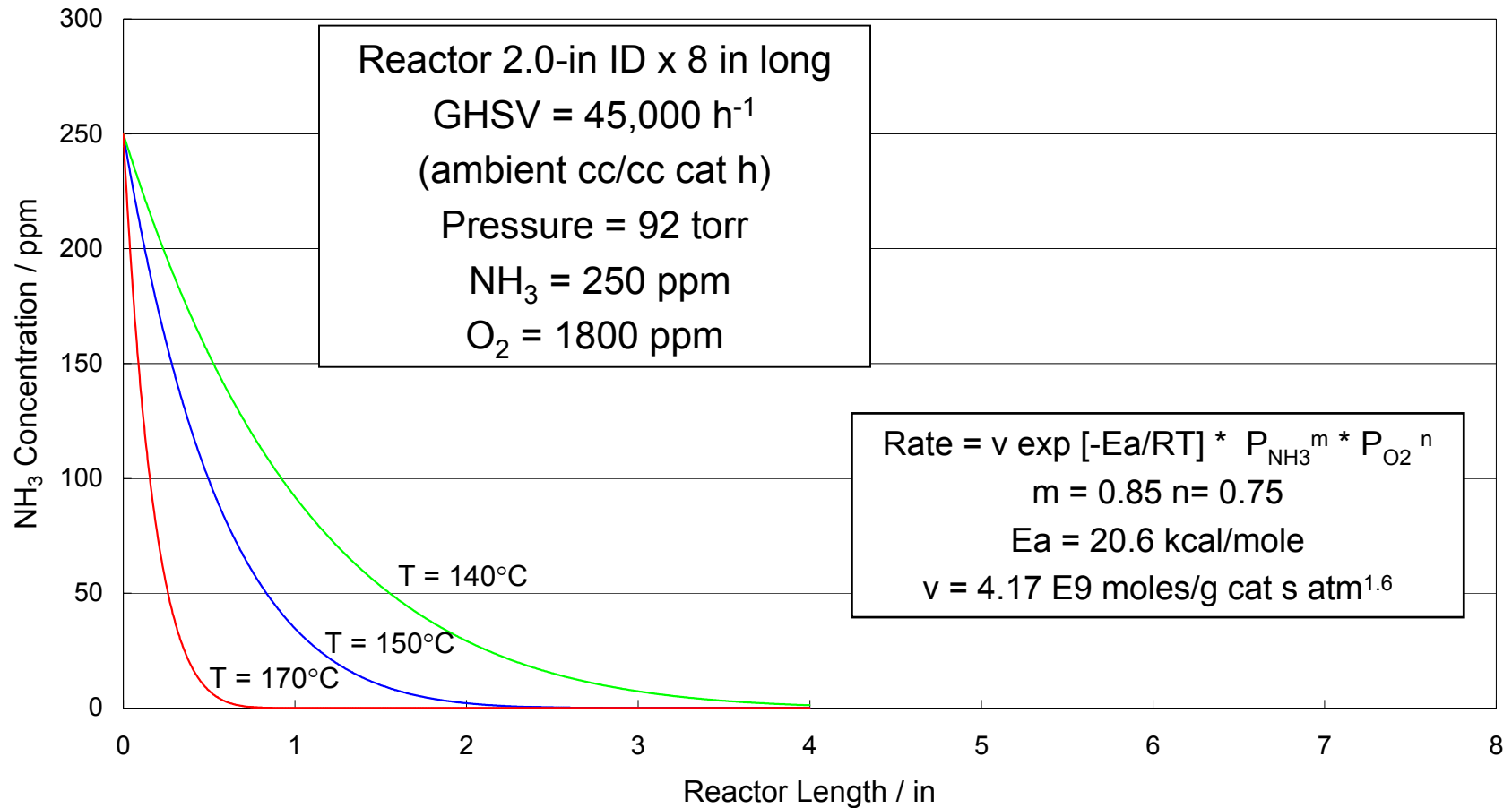
Pilot Scale Reactor



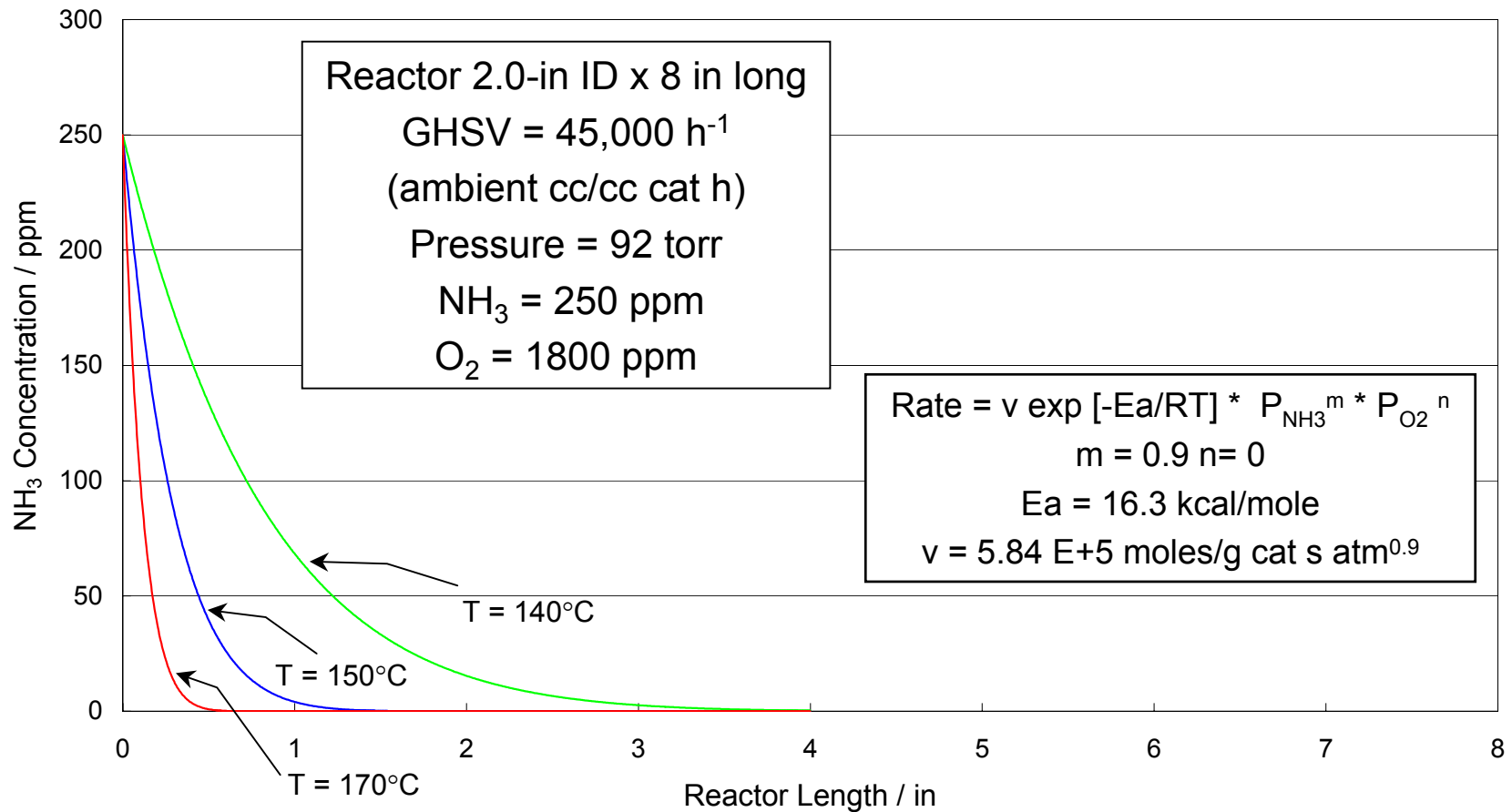
Catalyst Beds



Reactor Model Predicts High Conversion at $\leq 170^\circ\text{C}$



Reactor Model for HSSI Catalyst Also Predicts High Conversion



Hydrocarbon Oxidation

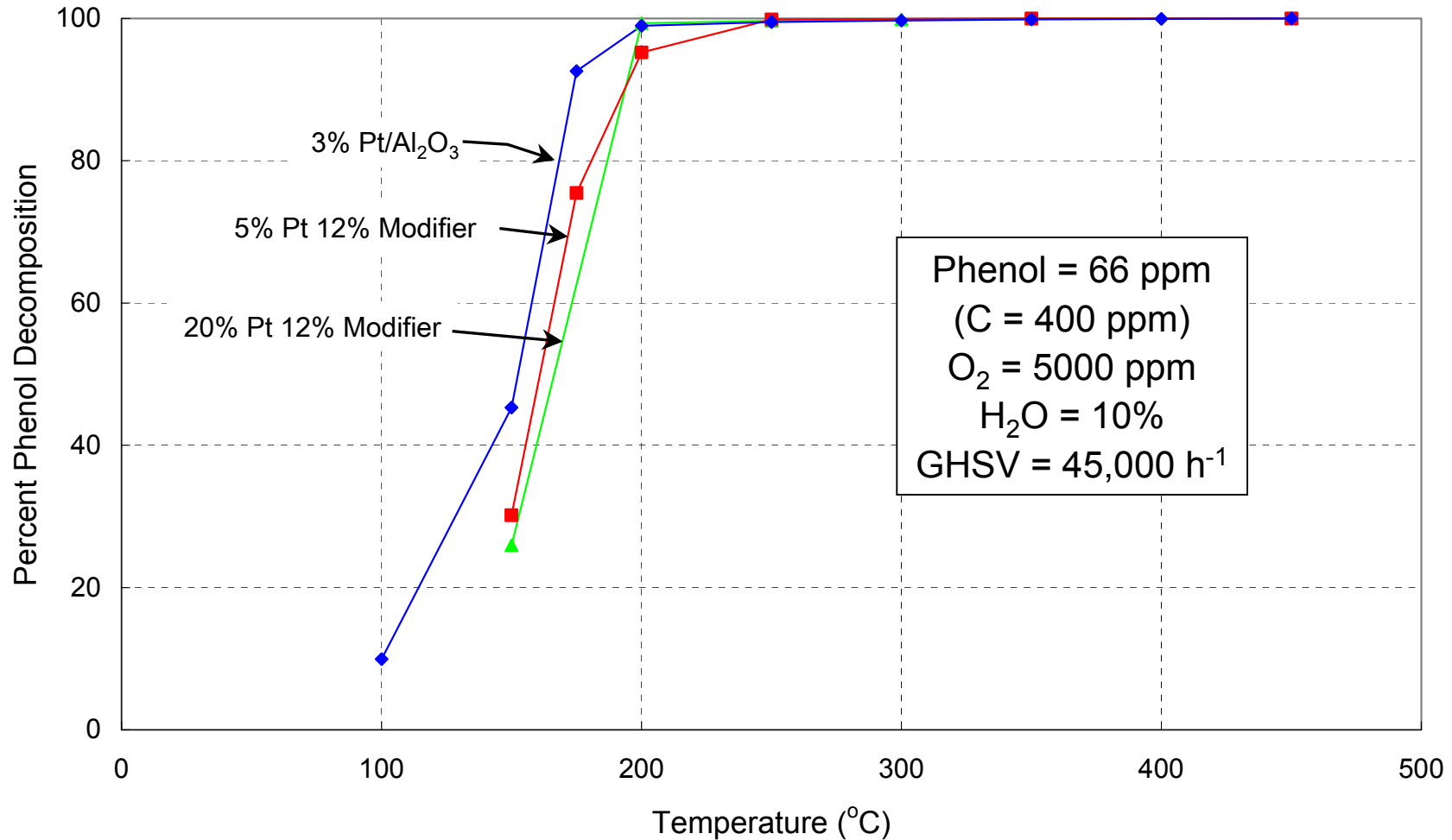
Ammonia oxidation occurs in the front end of the bed at low (<200°C).

Hydrocarbon oxidation will occur in the back end of the bed at higher temperature (400°C).

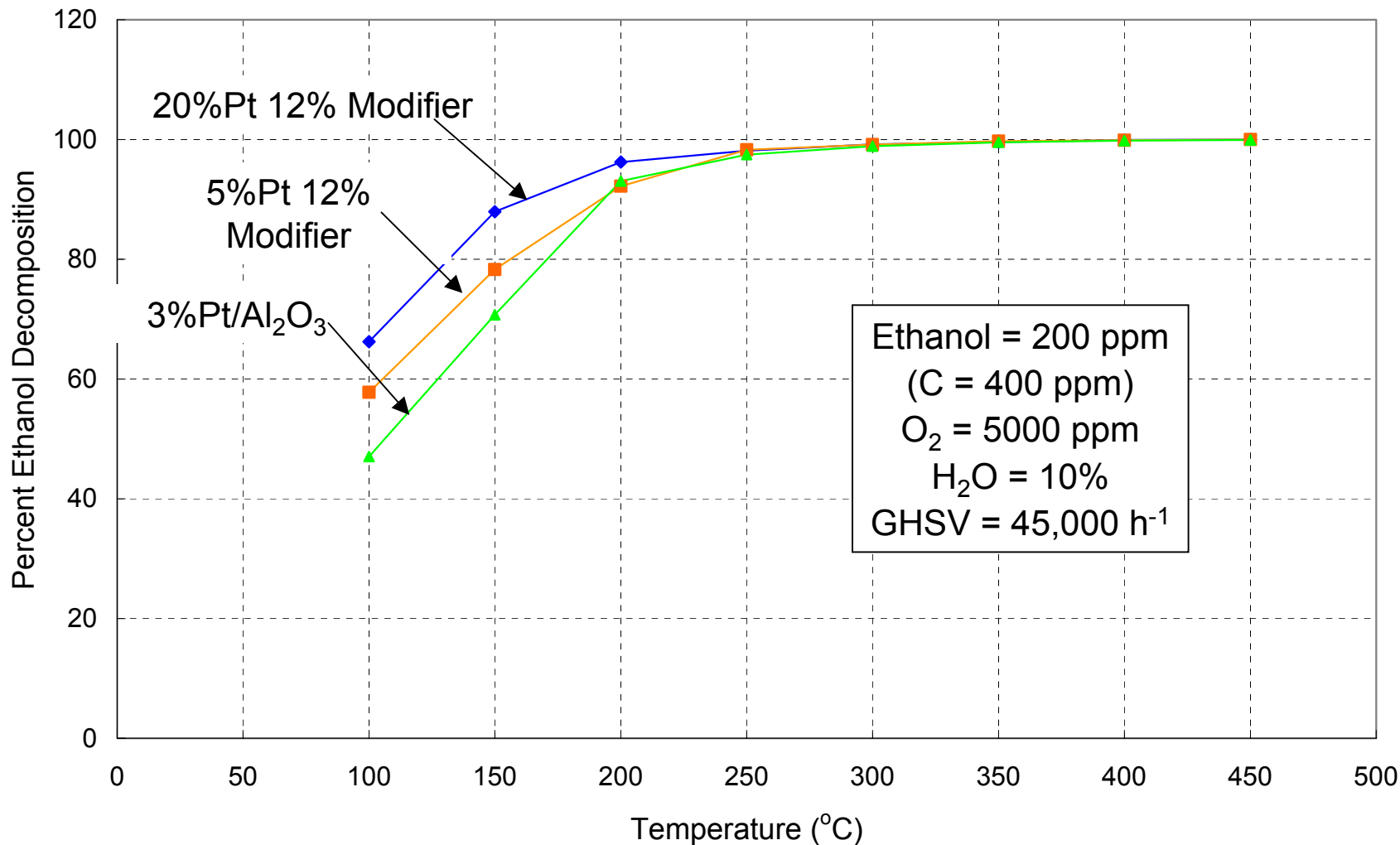
No NO_x is formed at 400°C, because NH_3 was converted to N_2 in the ammonia oxidation bed.

We need to determine what effect the unreacted hydrocarbons have on the ammonia oxidation bed.

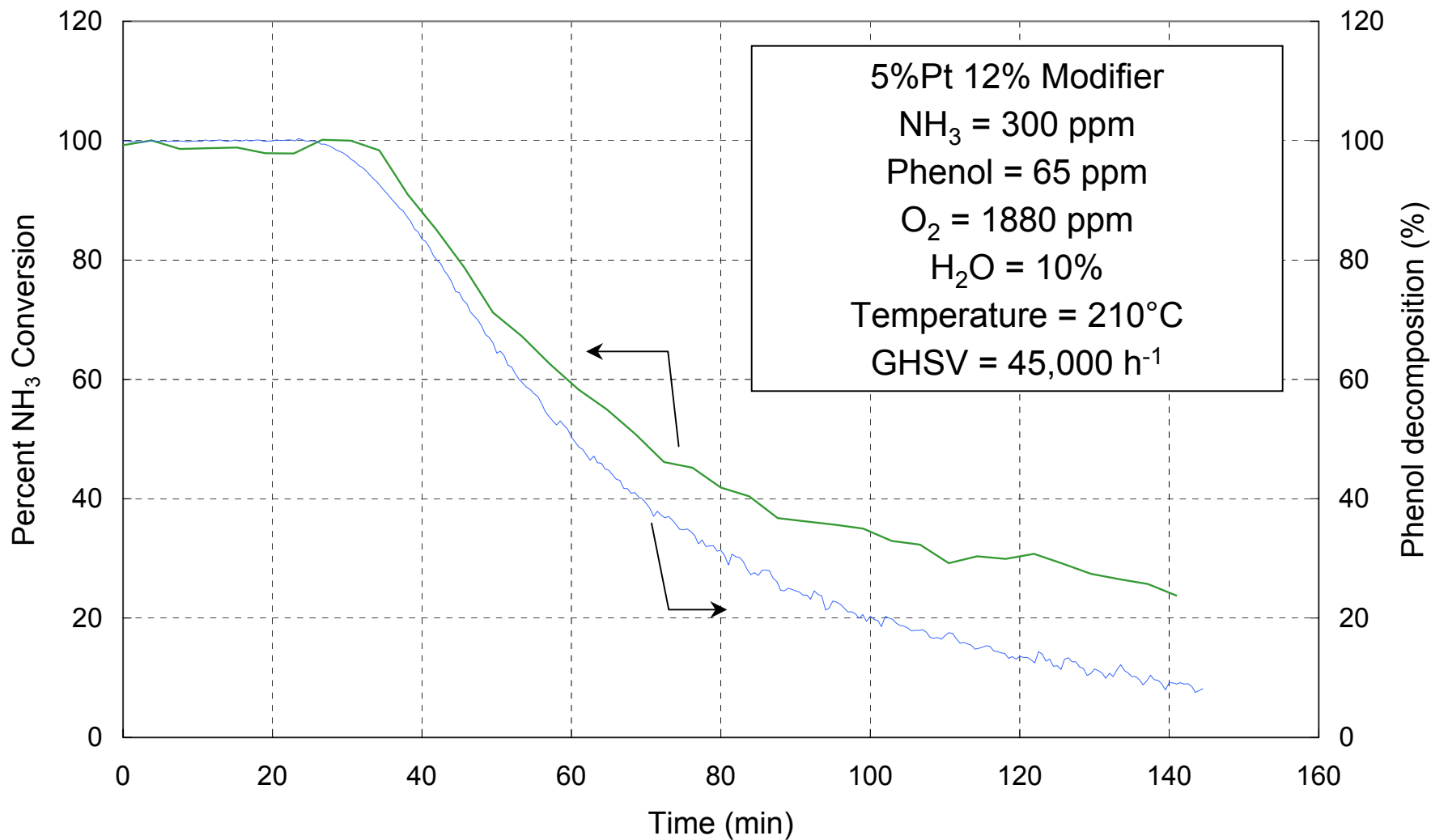
Oxidation of Aromatic Hydrocarbon is Complete by 200°C



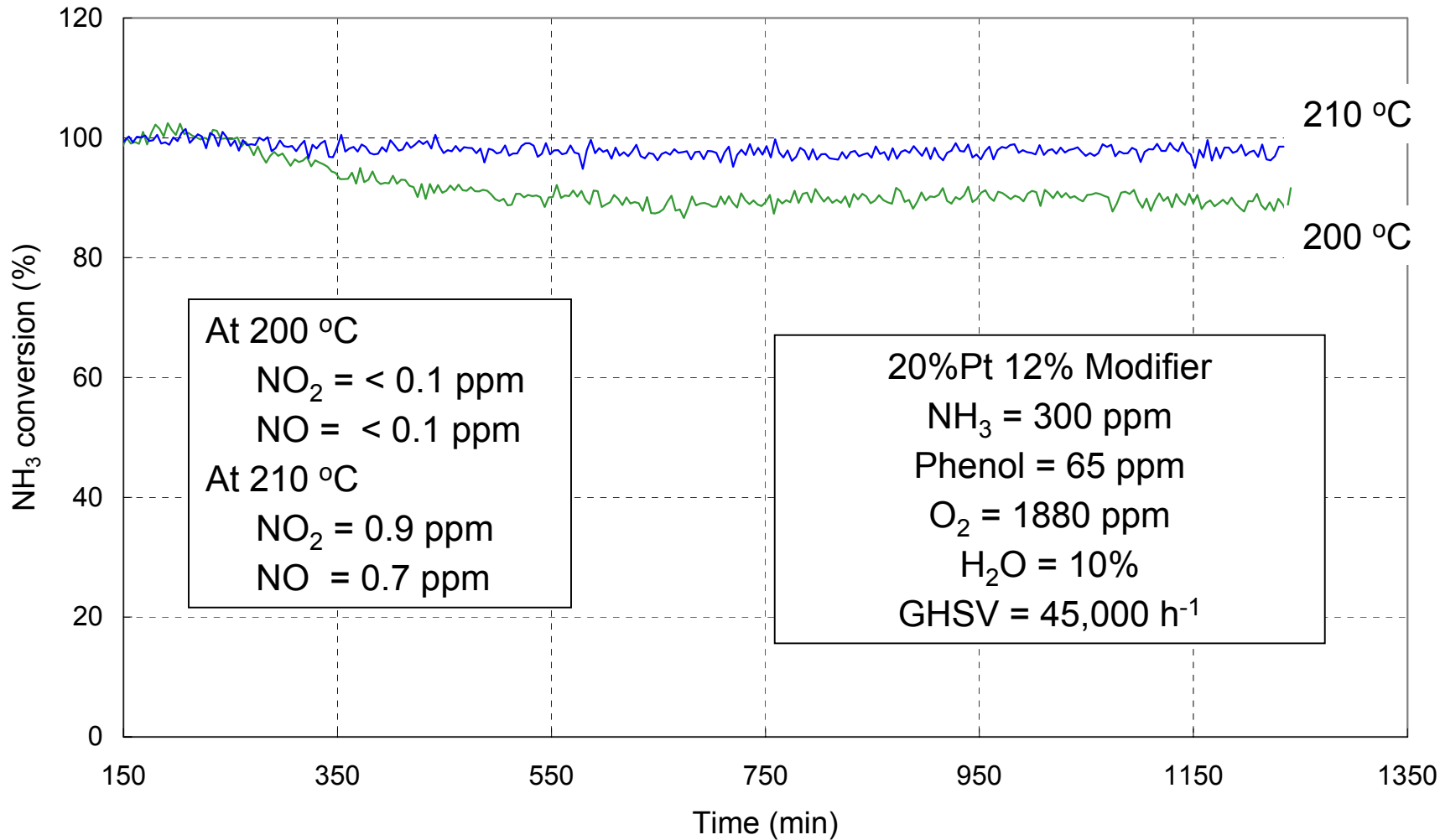
Catalysts are Effective for Oxidation of an Alcohol



Phenol Reduces Ammonia Oxidation



Higher Concentrations of Platinum Are Effective



Summary and Conclusions

TDA and HSSSI have identified catalysts formulations that are active for the oxidation of NO_x to H_2 and H_2O at 140°C under representative conditions.

The catalysts also produce very little NO_x at temperatures up to 200°C .

Our reactor model predicts high conversion half way through the pilot scale reactor at 150°C for both TDA and HSSSI catalysts.

We have identified catalysts that are suitable for HC oxidation.

Representative HCs may affect ammonia oxidation activity, but higher concentrations of platinum might help.

Acknowledgements

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John Fisher, COTR.