

NO oxidation and NO_x storage kinetics on Pt/Al₂O₃ and Pt/BaO/Al₂O₃

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Reference:

A kinetic study of NO oxidation and NO_x storage on Pt/Al₂O₃ and Pt/BaO/Al₂O₃, J. Phys. Chem. B, 2001, 105

Methodology :

Experiments were conducted using monolith reactor with 69 (1mm x 1mm) catalyzed channels. Reaction rates (pre-exponential factors, activation energies and coverage dependencies) are computed using laminar flow models.

Range of conditions :

Pressure : 1 atm (fixed)
Temperature : 300 C - 650 C NO_x concentrations : 0 - 700 ppm.
Space velocity : 87000 hr⁻¹

Experimental features :

Experiments were conducted using a monolith reactor with 69 (1mm x 1mm) channels of 15mm length. Ar and N₂ were used as inert gases. Effectiveness of Pt was reduced significantly (about 15 times for the catalysts used in these experiments) in the presence of BaO. Pre-exponential factors for adsorption were estimated using kinetic theory at a fixed temperature of 600 K (and their temperature dependence was neglected). Transition state theory is used to prevent unreasonable entropy changes. Catalyst is regenerated thermally without using a reductant.

Measurement techniques :

Pt site dispersions were measured using CO TPD by assuming maximum coverage of 0.7 CO molecule per Pt site. Dispersions were measured in a FTIR study of CO adsorbed on Pt when BaO was present. Pt dispersion

reduced by 5 times when BaO was present. Temperature measurements were made using thermocouples. Gases are detected using a mass spectrometer. NO and NO₂ were however measured using a chemiluminescence detector and N₂O was measured using an IR instrument.

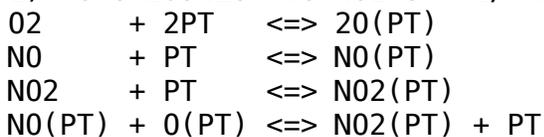
Materials/catalysts preparation :

In-house NO_x adsorber catalyst was prepared by applying an alumina-based washcoat onto a monolith. The specific procedure used is detailed in "A Kinetic Study of Oxygen Adsorption/Desorption and NO Oxidation over Pt/Al₂O₃ Catalysts," Louise Olsson, Björn Westerberg, Hans Persson, Erik Fridell, Magnus Skoglundh, and Bengt Andersson, J. Phys. Chem. B, 1999, 103.

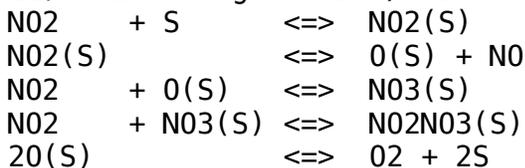
Detailed Description/Comments:

A NO_x storage and release cycle is assumed to consist of 4 steps.

i) NO oxidation to NO₂ on Pt/Al₂O₃.



ii) NO₂ storage on BaO/Al₂O₃



iii) NO_x release from BaO/Al₂O₃ (following an exotherm)



iv) NO_x reduction to N₂.

The files here (in a CHEMKIN format) provide the reaction mechanisms for the first 3 steps in the cycle.

Features of the mechanisms:

i) All reactions are reversible.

ii) All reactions have kinetic rates in the form of Arrhenius type expressions.

iii) Some reactions can have coverage dependent activation energies.

Effectiveness of Pt to oxidize NO is found to reduce in the presence of BaO. For the specific catalyst formulation (Pt and BaO particle loadings and their distributions) used in the experiments, effectiveness of Pt was reduced by 15 times. For lack of a sufficient

explanation or model equations, this "masking" effect, for now, has to be handled by multiplying the Pt loading with an effectiveness factor when BaO is present. The effectiveness factor would depend on BaO loading and some measure of the proximity between Pt and BaO sites. For catalyst loading comparable to those used in the experiments by Olsson et al., an effectiveness factor of 15 can be used.

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