Two-Nozzle Flame Synthesis of NO_x Storage- Reduction Catalysts

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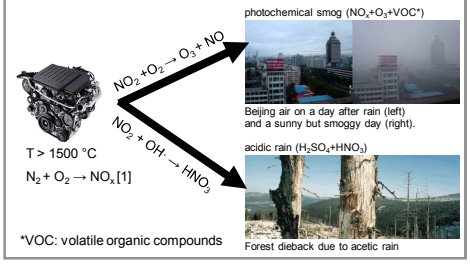
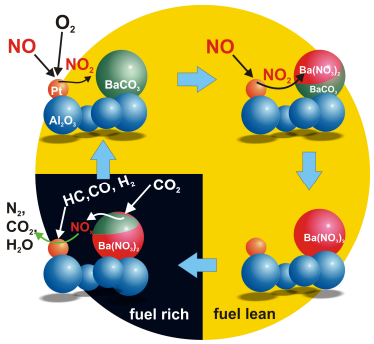
Objective

NO_x storage reduction (NSR) catalysts are used for abatement of combustion generated NO_x in lean burn engines. The effluent NO_x can be stored during the fuel lean phase on the Ba compound.



*VOC: volatile organic compounds

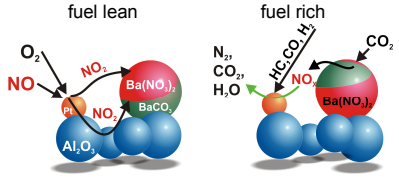
NO_x storage and reduction



Simplified NSR working principle: During fuel lean conditions NO_x is stored in the form of Ba-nitrates which is regenerated during fuel rich conditions.
Here, the influence of the Pt and Rh position on the NSR behavior is investigated by adapting the synthesis condition of the flame spray pyrolysis unit. Additionally the influence of SO_2, a catalysts poison, was investigated.

Noble metal - position

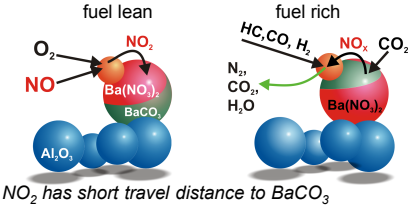
Pt on alumina support



NO_2 has to travel to the $BaCO_3$ sites

Pt on alumina is well known for good conversion of NO to NO_2 being transported over the gas or the surface to the Ba sites.

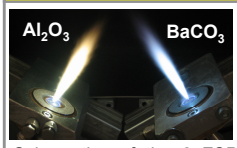
Pt on $BaCO_3$

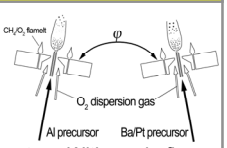


NO_2 has short travel distance to $BaCO_3$

In this case Pt is right on top of the $BaCO_3$ and NO_2 formation near to the Ba sites. Storage and especially reduction are facilitated because of the short transport distance.

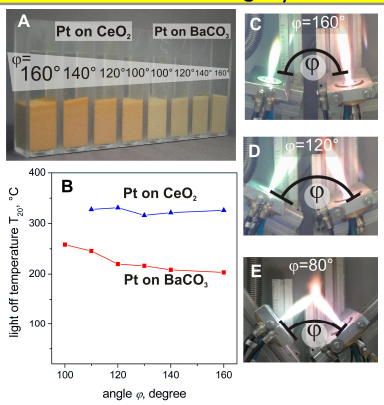
Particle production





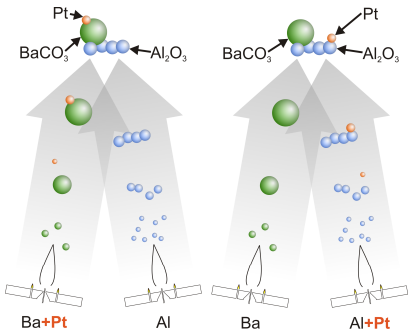
Schematics of the 2 FSP setup. With each flame particles are produced independently which mix continuously upstream of the flame.

Influence of angle φ



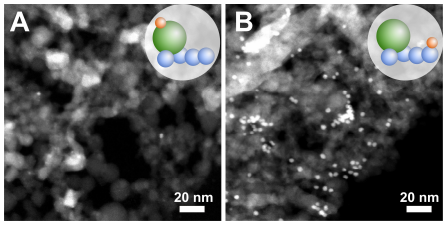
A) Color change as a function of the angle φ and Pt position and B) the corresponding light-off temperatures. The production under different angles changes the distance the two products are mixed C) φ=160° and D) φ=120° and E) φ=80°.

Selective noble metal position



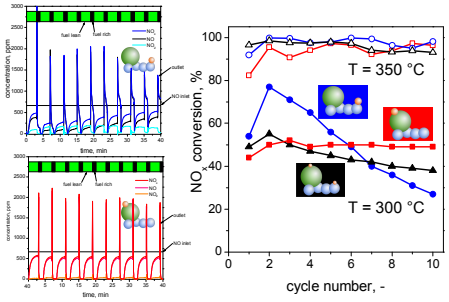
By adding Pt to the Ba-containing precursor (left side) preferential Pt deposition on $BaCO_3$ was made. Adding Pt to the Al-containing precursor (right side) resulted in preferential Pt deposition on Al_2O_3. [2]

Selective noble metal position

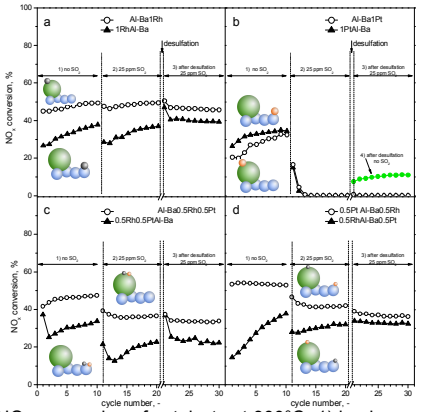


STEM image of Pt/Ba/Al_2O_3 catalysts with preferential Pt deposition. As Pt and Ba have about the same contrast they are difficult to see (A). Individual Pt on Al_2O_3 can be seen well in (B).

NSR performance



Outlet NO_x and NO_x conversion of preferentially located Pt. All three catalysts with the composition of Pt/Ba/Al_2O_3 = 1/20/100 weight ratio. Fuel lean period (3 min): 666 ppm NO and 3.3% O_2, balance He. Fuel rich period (1 min): 666 ppm NO and 1'333 ppm C_3H_6, balance He. Space velocity of 72'000 h^{-1}.



NO_x conversion of catalysts at 300°C: 1) in absence; 2) in presence of 25 ppm SO_2; 3) in presence of 25 ppm SO_2 after desulfation with 2% H_2 at 750 °C. Fuel lean period (5 min): 400 ppm NO, 500 ppm C_3H_6, and 8% O_2, balance He. Fuel rich period (5 min): 400 ppm NO and 500 ppm C_3H_6, balance He. Both at 300 °C and space velocity of 300'000 h^{-1}.

Conclusions

- With the 2-FSP setup nano-scale mixed powder can be produced, avoiding the formation of mixed oxides (atomic mixing).
- Noble metals, like Pt or Rh, can be preferentially deposited; here on storage or support sites.
- Bimetallic Rh-Pt showed high SO_2 tolerance and did not age at elevated temperatures up to 750°C.

Acknowledgements

We kindly acknowledge financial support by ETH Zürich (ETH Research Grant TH-09 06-2) and the European Research Council/European Community (under FP7). The contribution of platinum chemicals by Johnson Matthey PLC is greatly appreciated.

References

[1] Zeldovich, Acta physicochimica URSS, (1946) 577-628.
[2] R. Büchel, R. Strobel, A. Baiker, S.E. Pratsinis, Top. Catal. 52 (2009) 1709-1712.
[3] Büchel, R. Strobel, F. Krumeich, A. Baiker, S.E. Pratsinis, J. Catal. 261 (2009) 201-207.