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Gardini, Diego; Christiansen, J. M.; Jensen, Anker Degn; Damsgaard, Christian Danvad; Wagner, Jakob Birkedal

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Fundamental insight in soot oxidation over an Ag/Co$_3$O$_4$ catalyst by means of Environmental TEM

D. Gardini$^1$, J. M. Christiansen$^2$, A. Degn$^2$, C. D. Damsgaard$^{1,3}$, J. B. Wagner$^1$

$^1$ Center for Electron Nanoscopy, Technical University of Denmark, DK-2800 Lyngby, Denmark
$^2$ Department of Chemical and Biochemical Engineering, Technical University of Denmark, DK-2800 Lyngby, Denmark
$^3$ CINF, Department of Physics, Technical University of Denmark, DK-2800 Lyngby, Denmark

Motivation: Soot emitted from diesel engine as typical solid particles has caused acute health problems to human beings. In modern vehicles, soot filters need to be regenerated periodically through a catalyzed high temperature oxidation process involving an extra fuel consumption. A dream catalyst would oxidize soot at a very low ignition temperature $T_{ig}$, ideally the temperature of the exhaust gas itself ($T_{ig} < 250$ °C).

Choice of the materials:
Soot oxidation is a gas/solid/solid interaction. When catalyst and soot are crushed together (tight contact) the oxidation occurs at lower temperature, than when the two are stirred together with a spatula (loose contact).

Catalytic test of Ag/Co$_3$O$_4$, Ag and Co$_3$O$_4$:
Temperature programmed oxidation (TPO) in a flow reactor setup.
10-18 mg soot/catalyst mixture (ratio: 1/5 Wt/Wt).
1 NL/min flow of 10 vol% O$_2$ in N$_2$. 100 to 750 °C at a rate 11 °C/min.
CO and CO$_2$ concentrations in the effluent gas are measured with an ABB AO2020 continuous IR gas analyzer.

$\Rightarrow$ Performance of cosupported catalyst cannot be directly described in terms of the activity of the single Ag and Co$_3$O$_4$ components

Ex situ TEM analysis: TPO snapshots of Ag/soot system.
• Tapering of agglomerate’s borders at oxidation peak
• Soot consumption

In situ TEM analysis:
$T=350$ °C
• Initial soot consumption
• Ag starts to be mobile

$T=600$ °C
• Highly mobile Ag
• Soot gets consumed as Ag advances
• Diffraction halos are visible for the advancing Ag (crystallinity)

$T=750$ °C
• Soot oxidation is an exothermic process. Local heating could lead to melting of the advancing Ag (seen on metal nanoparticles in literature [1] for graphite oxidation). Once oxidation is over, Ag could solidify again.
• At higher temperature, Ag agglomerates back leaving a soot “snake skin” and forming the tapered fronts.

References:

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