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Identifying the transient phase in CO oxidation by XAS spectroscopy

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Supported platinum catalysts for the oxidation of carbon monoxide adopt different structures depending on temperature and reactant concentrations. These structures, which show vastly different activity, can be identified by X-ray absorption spectroscopy (XAS). While the low-temperature, low-activity state and the high-temperature, high-activity state are each well characterized, there is evidence of an additional transient phase, the nature of which is subject of strong debate[1].
Identifying a transient state poses a particular challenge, as both the transient phases'spectrum and its concentration are unknown, essentially turning the analysis into an optimization problem.
In this work, we investigate the nature of the transient state on platinum catalysts in CO oxidation through transient X-ray absorption spectroscopy. By switching the gas feed from pure CO to a CO oxygen mix at high temperature, light-off is induced and the change in the catalyst structure is followed by time resolved XAS. The intermediate state is shown to contain a significant amount of bare platinum surface.
Through linear combination fitting, time dependent concentrations of all involved species could be obtained. A reaction mechanism is proposed and it is shown that it offers a significant improvement in the fit of the concentrations compared to a simple two-step mechanism.

[1] a) R. van Rijn, O. Balmes, R. Felici, J. Gustafson, D. Wermeille, R. Westerström, E. Lundgren, J. W. M. Frenken, The Journal of Physical Chemistry C 2010, 114, 6875-6876; b) F. Gao, Y. Wang, D. W. Goodman, The Journal of Physical Chemistry C 2010, 114, 6874-6874.

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