Adventures in Catalytic Nanospace: imaging surface reactions at the atomic scale with scanning tunnelling microscopy

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2010年2月19日（金）14:45-15:45

Heterogeneous catalysis is usually accomplished with materials in nanoparticulate form. In this presentation I will describe one of our approaches to understanding catalytic processes and materials. This approach involves the fabrication of ultra-nanoparticulate model catalysts using MVD (metal vapour deposition) and MOCVD (metal-organic chemical vapour deposition) to form particles in the size range 1-10 nm. which can be imaged at very high spatial resolution (and in some cases at atomic resolution) using STM (scanning tunnelling microscopy). We have concentrated mainly on a system which we have used for anaerobic photocatalytic hydrogen production, namely Pd/TiO$_2$ powdered catalysts. Important catalytic phenomena resolved by these methods include:

1. **Spillover.** We have identified oxygen spillover from Pd nanoparticles supported on single crystal TiO$_2$ as a ring of material extending about 3nm. from the edge of the nanoparticles onto the support.
2. **SMSI.** The strong metal-support interaction is resolved at atomic resolution as an ‘alloyed’ surface layer of Pd:Ti with two main structures of 1:1 and 1:2 atomic ratio. The Ti is oxidised, but is probably only in the +2 state (by XPS).
3. **Oxygen Storage.** Growth of TiO$_2$ layers around the Pd nanoparticles occurs due to the conversion of Ti$^{3+}$ on the support to Ti$^{4+}$.
4. **The ‘Collection Zone’.** Using a molecular beam reactor we have shown that the adsorption of CO on Pd nanoparticles occurs with unexpectedly high sticking probability. This is due to the capture of CO into a weakly held, very short-lived state on the TiO$_2$ support, which, in its short sojourn, can nevertheless diffuse over a large number of sites, even at elevated temperature. In this way the molecules can ‘find’ the Pd nanoparticles where the CO is much more strongly bound.

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