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SMALL DEPOSITED METAL PARTICLES ON WELL-CHARACTERISED OXIDE SURFACES: MODELS FOR HETEROGENEOUS CATALYST

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Model supported metal catalysts have been prepared by deposition of transition metal vapour onto thin well-ordered oxide layers, in particular alumina [1–4]. The average sizes of the metal particles and their size distributions are determined by nucleation and growth. Here we review the morphology and structure of the oxide layers and the metal particles dispersed on them, as measured with a variety of methods including LEED and STM [5]. Electronic and magnetic structures as a function of particle size, adsorption properties, as well as reactivity with varying particle size is reviewed [6].

The electronic structure of supported Pd particles has been studied indicating a non-metal to metal transition occurring for particles exceeding a size of 70–80 atoms per aggregate. Adsorption of CO has been studied in detail applying FT-infrared spectroscopy. Interesting variations in the spectra when very small particles consisting only of a few metal atoms are investigated as compared with larger particles and single crystal surfaces are observed [7]. Ultra-small Rh, Pd, and Ir particles with minimum average sizes of five atoms have been grown at temperatures of 90 K and below. In the infrared spectra of adsorbed carbon monoxide, characteristic features were observed by infrared reflection absorption spectroscopy (IRAS). These originate from CO adsorbed on single metal atoms and extremely small aggregates, most likely dimers or trimers. Located at oxide point defects, rhodium atoms form Rh dicarbonyl species with a symmetric stretching frequency of 2117 cm^{-1} , while for Ir dicarbonyl species a frequency of 2107 cm^{-1} was found. At 60 K, Rh aggregates also grow elsewhere on the oxide film. Comparing the nucleation behaviour at low temperatures, we find varying metal-oxide interaction strength for different transition metals.

CO dissociation has been studied on Rh aggregates and if a maximal dissociation rate has been found for aggregates containing several hundred metal atoms [8]. The presence of defects on the particles is deduced to be origin of this behaviour.

While these studies have been performed under UHV conditions, it has now become possible to perform *in situ* spectroscopy under ambient conditions.

Infrared-visible sum frequency generation (SFG) vibrational spectroscopy is one of the few surface-specific techniques that can operate in a pressure range from ultrahigh vacuum to ambient conditions. Due to its inherent surface sensitivity and pressure independence, SFG is particularly suited for *in situ* studies of adsorbates or surface species at elevated pressure or during a catalytic reaction [9–11]. We have designed an SFG-compatible elevated pressure reactor that is attached to an ultrahigh vacuum (UHV) surface analysis chamber. After preparation and characterisation in UHV, model catalysts can be transferred in vacuo into the reaction cell. The reactor is separated from the UHV chamber by an arrangement of differentially pumped Teflon seals.

We present results on CO adsorbates on the above described and characterised model catalysts [12,13].

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