

CO Oxidation over IrO₂(110) – A DFT Study

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IrO₂ shows good performance for a range of reactions in heterogeneous catalysis. Amongst others, it is one of the most active materials for electrochemical water oxidation [1] and displays a very low barrier for methane dissociation [2]. Despite these important applications, studies on Ir oxide are mostly limited to its electrochemical properties [1] and the oxide growth upon oxidation of Ir(111) [3]. Initial studies indicate, that oxidation of Ir to IrO₂ only proceeds at elevated temperatures and comparably high oxygen partial pressures [3]. Under these circumstances a fully oxidized IrO₂(110) surface must be expected [3].

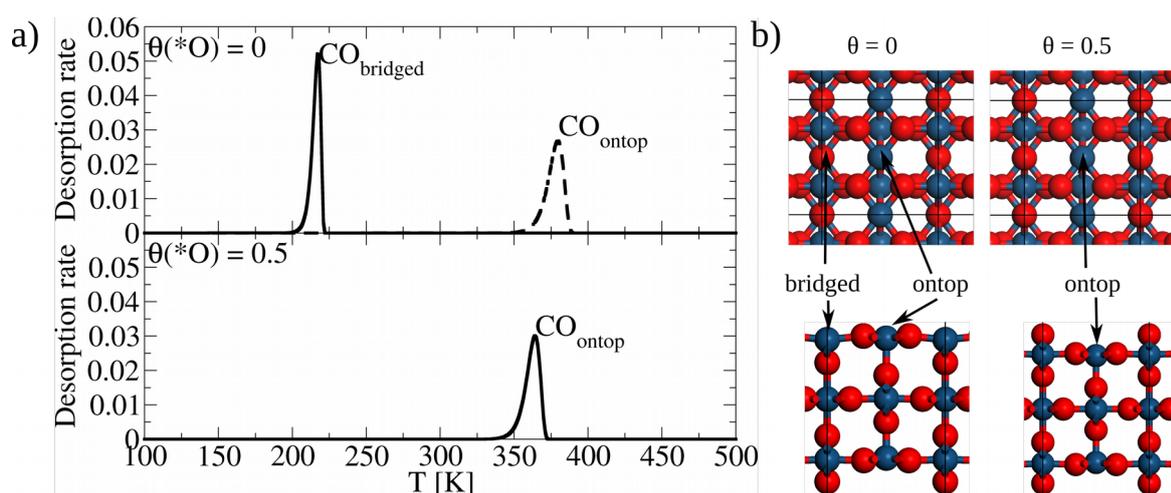


Figure 1: a) Simulated temperature programmed desorption (TPD) spectrum for CO adsorption at IrO₂(110). b) Model systems of the IrO₂(110) surface.

To close this gap in the understanding of the chemistry of the IrO₂(110) surface, we have studied CO adsorption and oxidation as model reactions. Initial experiments indicate, that CO adsorption is not possible at the freshly prepared IrO₂(110) surface. Instead, CO adsorbs only after annealing. These experiments are corroborated by density functional theory (DFT) calculations which indicate, that CO binds strongly to the ontop sites (Figure 1). Furthermore, the adsorption strength of CO is not affected by the presence of oxygen co-adsorbates (Figure 1). Thus, CO adsorption should be possible provided free adsorption sites are available.

To understand the possible effect of the flash annealing, we have explored the surface configuration of IrO₂(110) under H₂O and O₂ exposure and the influence of elevated temperatures by performing ab initio thermodynamics. Knowing the state of the surface, we have investigated CO oxidation. The results are compared to CO adsorption and oxidation over other transition metal oxides.

References:

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