Interaction of bimetallic Zn/Au(111) surfaces with O_2 or NO_2 and formation of ZnO_x/Au(111)

Konstantin M. Schüttler, Joachim Bansmann, Albert K. Engstfeld[+] and R. Jürgen Behm*

Institute of Surface Chemistry and Catalysis, Ulm University,

Albert-Einstein-Allee 47, D-89081 Ulm, Germany

Abstract

Continuing our efforts to elucidate the working principle of Au/ZnO catalysts for the synthesis of green methanol from CO_2 and H_2, we present a systematic X-ray photoelectron spectroscopy (XPS) study on the interaction of bimetallic Zn/Au(111) surfaces with O_2 and NO_2 (O^*), on the oxidation of Zn atoms in these bimetallic model systems and on the formation ZnO_x species or thin films on these surfaces. At room temperature, we find little adsorption and essentially no oxidation of Zn atoms alloyed into the Au(111) near-surface region upon exposure to O_2, and accordingly no Zn de-alloying. LT interaction of O_2 or NO_2 with Zn submonolayer structures leads to higher oxygen and/or NO_2 coverages. Formation of oxidic ZnO_x species / structures occurs, however, only after annealing above RT, both after O_2 and NO_2 exposure. This involves partial conversion of adsorbed species into ZnO_x. ZnO/Au(111) thin films, especially thicker ones, can be efficiently prepared by reactive deposition of Zn in O_2 or NO_2, followed by annealing. RT deposition in NO_2 gives a higher fraction of (incompletely) oxidized ZnO_x compared to O_2, Fully oxidized ZnO is formed after subsequent annealing, upon thermally activated conversion of adsorbed O_{ad}/OH species into oxidic oxygen and dissolution of remaining metallic Zn atoms into the Au bulk. Overall, the study provides detailed insights into the formation of ZnO_x thin films on Au(111).

Keywords: *Bimetallic Surface, Oxide film formation, Reactive deposition, De-intermixing; Zn/Au(111), ZnO/Au(111), X-Ray Photoelectron Spectroscopy (XPS)*

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+ Pres. address: Institute of Electrochemistry, Ulm University
* Author, to whom correspondence should be sent, e-mail: juergen.behm@uni-ulm.de