Adsorption, reaction and desorption of hydrogen on modified Pd(1 1 1) surfaces

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ABSTRACT

The interaction of hydrogen (deuterium) with different modified Pd(111) surfaces has been investigated. The focus was put on the energy and angular dependence of the desorbing molecules from oxygen covered, potassium covered and vanadium oxide covered surfaces. Conventional adsorption/desorption as well as permeation/desorption experiments were performed. For the oxygen covered surface optimum reaction rates for water production and the energy distribution of the reaction products were determined, both for the reaction of oxygen with molecular hydrogen as well as with atomic hydrogen. Potassium on the surface enhances the activation barrier for hydrogen adsorption resulting in a hyper-thermal desorption flux and a forward focused angular distribution of desorption. Permeation/desorption of deuterium from ultra-thin vanadium oxide films yield mainly thermalized desorbing molecules or slightly hyper-thermal, depending on the oxidation state of the surface oxide.

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1. Introduction

The interaction of hydrogen (deuterium) with solids is an issue which has attracted renewed interest in the recent years due to the promising application in a future hydrogen economy [1]. Hydrogen production by catalytic processes [2], hydrogen storage in solids [3] and hydrogen burning in fuel cells [4] are just some of the important technologies in this context. For all these applications the interaction of hydrogen with the solid surface plays a dominant role. It is now generally acknowledged that the efficient use of hydrogen to solve the energy problem requires a thorough understanding of all basic processes involved in hydrogen production, storage, cleaning and cold combustion. In this context palladium is a very attractive model material since it can easily adsorb hydrogen dissociatively and absorb large amounts of hydrogen in the bulk [5]. However, poisoning of the palladium surface by co-adsorbates can severely reduce the uptake of hydrogen.

In this article an overview on fundamental interaction processes for the hydrogen-palladium system, with special emphasis of the influence of chemical and structural modifications on a Pd(1 1 1) single crystal surface will be given. In particular, the kinetics and dynamics of adsorption and desorption will be addressed. The main focus is put on the kinetic energy and angle dependence of the hydrogen (deuterium) desorption flux, as well as of the reaction product water, which yields information on the activation barriers involved in the adsorption/desorption and reaction processes. With respect to the surface modifications the influence of oxygen, potassium and vanadium oxide on the interaction with hydrogen will be discussed. All the presented studies were performed under well controlled, ultra-high vacuum conditions, allowing a detailed characterisation of the surface conditions.

2. Experimental

The experiments were carried out in several ultra-high vacuum chambers with typical base pressures of $1 \times 10^{-10}$ mbar. The common surface analytical tools, as Auger electron spectroscopy (AES) and low energy electron diffraction (LEED) were applied to characterise the surface chemical composition and the surface structure, respectively. Typically, hydrogen was supplied in molecular form, but for some studies also atomic hydrogen was used for dosing. For this purpose a special doser for atomic hydrogen was used as described in detail elsewhere [6]. It basically consists of a tungsten tube which can be heated by electron bombardment up to 2000 K. In this case hydrogen flowing through the tube is dissociated to nearly 100%.

In most cases a Pd(1 1 1) sample with a diameter of 10 mm and 1.5 mm thickness was used, which was suspended on Ta wires.

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