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Kinetics of Interaction of Hydrogen with Nanostructured C–Pd Films for Hydrogen Sensing

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In this paper we describe the properties of nanostructural carbonaceous–palladium (C–Pd) films for hydrogen sensing. These C–Pd films were prepared by Physical Vapor Deposition method. The structure and morphology of the obtained films were characterized by SEM and TEM techniques and then hydrogen sensing measurements were performed. The films are composed of palladium nanograins placed in carbonaceous matrix. The size of palladium nanograins is ∼10 nm. The sensitivity and response rate of the films to hydrogen presence is proportional to square root of H_2 partial pressure in the ambient atmosphere. The dissociation of H_2 molecules on the surface of palladium nanograins is the rate-limiting step of hydrogen absorption.

Keywords: Hydrogen Sensor, C–Pd Film, Hydrogen Absorption.

RESEARCH ARTICLE

1. INTRODUCTION

Development and expanded use of hydrogen gas as an energy carrier caused the increasing demand for fast and reliable hydrogen sensors. It is due to high flammability and explosiveness of hydrogen when its concentration exceeds 4% in air.[1] Palladium-based hydrogen sensors have been thoroughly explored because of highly selective interaction between palladium and hydrogen.[2] The sensing mechanism of these sensors is based on the resistance changes of Pd in the presence of hydrogen which can be linked with H_2 concentration. Interaction between hydrogen and palladium begins with adsorption of H_2 on palladium surface, following homolytic dissociation of hydrogen molecules to H atoms. These hydrogen atoms diffuse into Pd lattice and occupy its interstitial sites, forming solid solution.[2,3] At higher hydrogen pressure (above 1–2 kPa), further incorporation of hydrogen atoms induces a phase transition from α- to β-phase and creation of palladium hydride.[4,5] The Pd–H system is characterized by higher resistance than metallic palladium.[6]

The palladium-based sensors include palladium thin films,[4] Pd nanowires,[3,7] Pd nanoparticle layers,[8–10] as well as C–Pd composites.[11–13] The sensitivity and response time of various nanostructures are summarized in Table I. Palladium thin films are the easiest nanostructures to fabricate. However, Pd thin films suffer from hysteretic resistance behavior and structural deformations.[2] Palladium nanowires show similar sensor properties as Pd thin films, and even better in terms of response time.[3,4,7] The problem lies in difficulty in fabrication of these nanostructures.[2] Pd nanoparticles are characterized by a similar sensitivity as Pd thin films and nanowires, while their response time is significantly longer.

C–Pd composites sensors may be in the form of multi-walled carbon nanotubes (MWCNTs) decorated with palladium nanoparticles,[11] graphene decorated with palladium nanoparticles[12] or palladium nanoparticles embedded in carbonaceous matrix.[13] System based on MWCNTs is characterized by low sensitivity and very slow recovery.[11] Graphene decorated with Pd nanoparticles shows very high sensitivity. The problem is decreasing sensor response during the repeated H_2 sensing cycles, very slow response time as well as slow and incomplete recovery.[12] Therefore, nanostructural carbonaceous–palladium (C–Pd) films with palladium nanograins placed in porous carbon matrix seem to be promising composite materials for hydrogen sensor applications.[14]

In this paper we present the results of studies of interaction between hydrogen and C–Pd films. We found that the sensitivity and response rate of C–Pd films increased linearly with square root of H_2 partial pressure in the surrounding atmosphere. We also conclude from the obtained

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