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H_2 Gas Sensor Based on PdO_x doped In_2O_3 Synthesized by Flame Spray Pyrolysis

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Abstract:

0–1.0wt% Pd doped In_2O_3 nanoparticles have been successfully synthesized in a single step by flame spray pyrolysis (FSP) technique using Indium (III) nitrate hydrate and palladium (II) acetylacetonate, as precursors dissolved in ethanol and their hydrogen sensing characteristics have been investigated. The particle and sensing film properties were analyzed by XRD, BET, TEM and XPS. The sensing films were prepared by spin coating technique. The crystallite sizes of In_2O_3 spherical and cubic morphologies were found to be ranging from 2 to 20 nm and Pd might form solid solution with In_2O_3 lattice. Gas-sensing characterization were studied at the operating temperatures ranging from 150 to 350°C in dry air. Hydrogen sensing characteristics of In_2O_3 nanoparticles was significantly improved as Pd content increased from 0 to 1.0wt%. The 0.50wt%Pd doped In_2O_3 sensing film showed an optimum H_2 response of 3,526 towards 1.0vol% H_2 concentration at 250°C operating temperature. In addition, PdO_x doped In_2O_3 sensing films exhibited good selectivity towards hydrogen with improving of response and response time.

Key words: Flame spray pyrolysis, PdO_x doped In_2O_3, Gas sensor, H_2 sensor

Introduction

Hydrogen gas is expected to become a green and renewable energy source in the future for several applications, including fuel cell vehicles, space crafts, automobiles, power generators and aircrafts [1]. This light and odorless gas is highly flammable; hydrogen gas leaks can result in disastrous consequences, such as explosion [2]. However, H_2 gas is highly flammable and explosive at volume concentration higher than 4% mixed with air, thus leading to growing demands for high performance H_2 sensors to detect its leakage [3]. In this study the Pd doped In_2O_3 nanoparticles were synthesized by flame spray pyrolysis. The effect of Pd on gas sensing performances were systematically studied and optimized for selective detection of H_2 gas. At the same time, the range of interest for its detection is 150–10,000 ppm, allowing for early leakage warning and explosive indication. So it's essential to detect the content of H_2 to avoid more serious accidents at incipient faults.

Materials and Methods

Firstly, 0–1.0wt%Pd doped In_2O_3 nanoparticles were synthesized by flame spray pyrolysis, which was previously established by our group. For sensor fabrication, flame-made 0–1.0wt%Pd doped In_2O_3 nanopowders were thoroughly mixed and ground with the binder solution. The resulting paste was spin coated on Al_2O_3 substrates equipped with Au interdigitated electrodes to form a sensing film. The resulting substrates were annealed at 450°C for 2 h in an oven for binder removal prior to sensing test. The gas-sensing performances of all sensors were characterized towards H_2, C_2H_2, C_2H_4, C_2H_4O, C_2H_5OH, H_2S and NO_2 under atmospheric conditions by the standard flow through technique in stainless steel chamber at operate temperature in the range of 150–350°C.

Results and discussion

Characterizations significantly confirmed Pd^{2+} and Pd^{4+} was formed solid solution with In_2O_3

lattice. Fig.1 shows TEM images with corresponding SAED pattern of pure In_2O_3 and optimal Pd doped In_2O_3 and highest Pd doping. It is seen that the spherical and cubic nanoparticles with the particle size were in the range of 2–20 nm. The corresponding SAED patterns display diffraction rings of polycrystalline In_2O_3 nanoparticles having (211), (222), (400), (440) and (622) dominant planes in good agreement with the preferred crystallographic orientations previously seen from the XRD patterns while there was no plane associated with PdO or PdO_2 phases for Pd-doped In_2O_3 nanoparticles. The gas sensing characteristics were showed in Figures 2–5.The effect of operating temperature ranging from 150 to 350°C towards H_2 response of In_2O_3 nanoparticles with different Pd doping indicated that the 0.5 wt% PdO_x doped In_2O_3 sensor has the highest response at the optimum working temperature of 250°C exhibited high response of ~3,526 to 1.0vol% H_2.The response and recovery time were 2 s and 12 min respectively. The PdO_x doped In_2O_3 can improve sensor response, and decrease response and recovery time of sensor and exhibited good selectivity towards hydrogen gas.

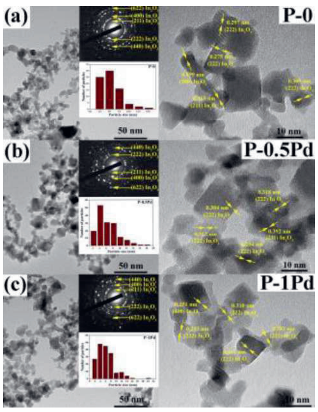

Fig.1. TEM images of the FSP-made (a) undoped In_2O_3, (b) 0.50wt% Pd-doped In_2O_3 and (c) 1.0wt%Pd-doped In_2O_3 nanoparticles. Insets: the corresponding SAED patterns and particle size distributions

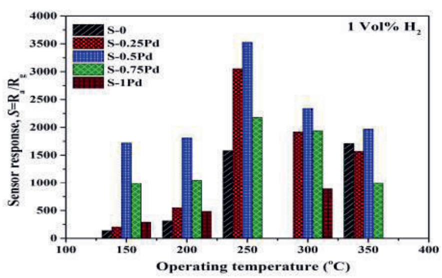

Fig.2. Histograms of sensor response to 1.0vol% of H_2 of FSP-made Pd-doped In_2O_3 sensors with different Pd contents (S-0 to S-1Pd) at various operating temperatures ranging from 150 to 350°C.

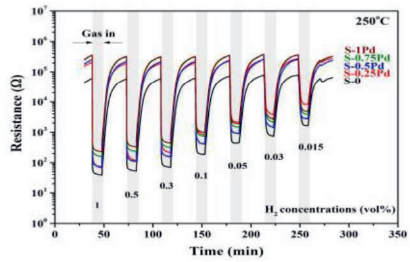

Fig.3. The change in resistance of In_2O_3 sensing films with different Pd doping concentrations under exposure to various H_2 concentrations ranging from 0.015 to 1.0vol% at the operating temperature of 250°C.

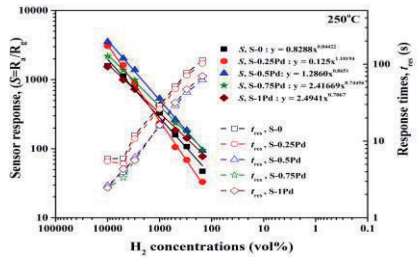

Fig.4. The corresponding sensor response(S) (left axis) and response time (t_{res}) (right axis) vs. H_2 concentration of 0–1.0wt%Pd doped In_2O_3 sensors at 250°C.

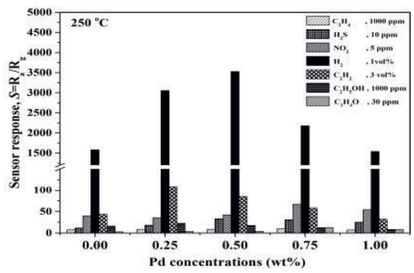

Fig.5. Selectivity histograms: sensor response of 0–1.0 wt% Pd doped In_2O_3 sensors to 1,000 ppm C_2H_4, 1,000 ppm C_2H_5OH, 3.0vol% C_2H_2, 1.0vol% H_2, 10 ppm H_2S and 5 ppm NO_2 at the optimal operating temperatures of 250°C.

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