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One step synthesis of Au loaded nanocrystalline SnO_2 by flame spray pyrolysis technique and its stability as gas sensor material

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Abstract:
One step synthesis of Au-loaded SnO_2-based nanocomposites for chemical gas sensors is presented. Flame spray pyrolysis (FSP) procedure with the use of originally designed setup was utilised to obtain either pure or Au-loaded materials. Superior gas sensor properties of obtained SnO_2-Au nanocomposites compared to same way prepared pure SnO_2 material are demonstrated. HAADF STEM microscopy was utilized to provide chemical insight into a degradation of gas sensor performance of given materials over time.

Key words: metal oxide, semiconductor, gas sensor, flame spray pyrolysis, tin dioxide, gold

Introduction

Flame spray pyrolysis procedure is currently considered to be a very promising route to obtain ultrafine porous materials on the basis of metal oxide semiconductors for chemical gas sensors [1]. The main benefits of this synthetic method among others are single synthetic stage, simplicity of precursor preparation, high rate of synthesis, good control of final material grain size, specific surface are, impurity content and homogeneity of modifier distribution [2]. This combination usually gives rise to a superior sensor performance of such obtained materials compared to samples obtained by convention multistage sol gel and impregnation techniques [3]. Gold modified nanocrystalline SnO2 is a well described material for semiconductor chemical gas sensor for its high sensitivity for reducing gases [4]. However, it is usually obtained by impregnation of nanocrystalline SnO2 with such precursors as water solutions of chloroauric acid or its salts with consecutive thermal decomposition [5,6]. This procedure has significant drawback as the final material gets contaminated with the Cl- or alkali metals ions. This can affect the materials sensitivity or long-term stability during gas sensor operation. Thus, a method of one step synthesis of SnO2-based Au loaded nanocomposites without contamination by other precursor component is of a high demand. Here we present this method, based on the use of Au-precursor soluble in organic solvent.

Experimental

Tin 2-ethylhexanoate has been taken as SnO_2 precursor, and (1,3-bis(2,6-diisopropylphenyl)-1,3-diazepan-2-ylidene)gold(I) as a precursor for gold. The SnO_2 precursor was mixed with toluene in 1:4 volume ratio and gold precursor was added to the solution in a calculated amount to give 0.05, 0.1, 0.2 and 0.4 % wt in the final material. The mixture was supplied with the 2ml/min rate and atomized through spraying nozzle by 10 l/min flow of oxygen and was ignited by the flame of six concentric orifices, supported by 1:2 methane to oxygen mixture by volume. Gold free material was synthesized in the same conditions. The materials were passed through characterization via XRD, BET, TPR-H_2, HAADF STEM and gas sensor test routine.

Results

Gas sensor response of synthesized materials towards 10 ppm of CO in dry air is given on the fig.1. The deterioration of sensors performance can be clearly observed for Au-modified materials during continuous work for 1 week in 50-450 working temperature range. Application of HAADF STEM analysis technique with EDX element mapping (Fig.2) allows to connect the observed materials performance drop to

increase of Au particle size from 2-3 to 15-20 nm, which causes a loss of their catalytic effect.

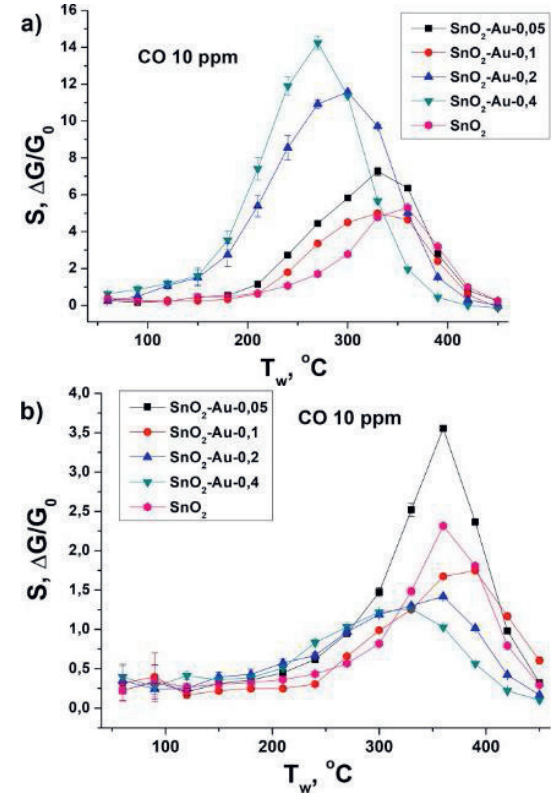

Fig. 1 Gas sensor response of obtained materials towards 10 ppm of CO in dry air a) after gas sensor assembly and b) after 1 week of continuous operation.

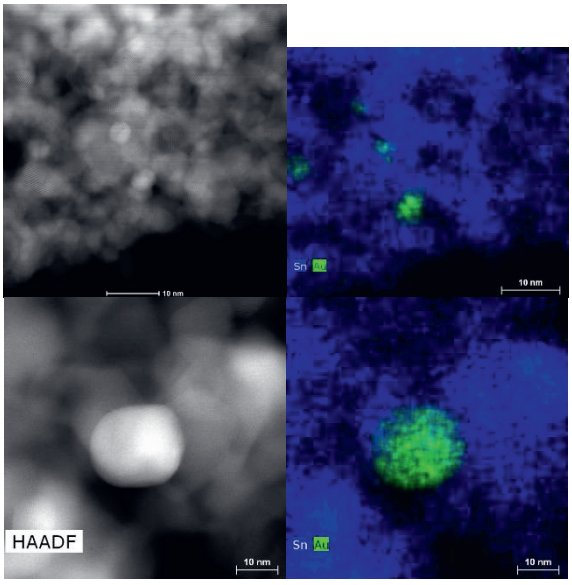

Fig.2 TEM micrograph with Sn and Au EDX mapping for as prepared SnO2-Au-0,2 material (top left and right) and after annealing at 500 °C for 24h in air (bottom left and right).

Conclusions

The FSP technique is a promising approach for the synthesis of gold modified SnO_2 based nanocomposites for gas sensors, however, the agglomeration of gold nanoparticles at elevated working temperatures leads to a complete deterioration of nanocomposites gas sensor performance. Stabilization of gold species required in order to achieve stable gas sensor response.

Acknowledgements

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