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Au- and Ni-promoted Ce-Zr catalytic filter-SnO_2 integrated sensor for dual selective detection of ethanol and methane

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Abstract

Gold and nickel nanoparticles on the $Ce_{0.8}Zr_{0.2}O_2$ mixed oxide are used as a catalytic filters for selective detection of ethanol and methane in presence of CO. Thick film SnO_2 gas sensor was covered with Au/Ce-Zr or Ni/Ce-Zr thick layers, to oxidize either CO or ethanol at different temperatures. A flow system was used to measure the sensor response to 300 ppm carbon monoxide or ethanol and 5000 ppm methane at 100-480ºC. Without using the filter, the SnO_2 sensor shows comparable responses to carbon monoxide and methane and much higher ones to ethanol. The Au/Ce-Zr catalytic filter completely converts carbon monoxide and ethanol to the insensitive CO_2 at 200 and 350ºC, respectively. The corresponding temperatures for Ni/Ce-Zr catalytic filter are 380 and 410ºC. No significant conversion of methane is observed at 100-480ºC on none of the filters. The operating temperatures of the sensor covered with Au/Ce-Zr filter for selective detection of ethanol and methane are 200°C and higher than 350°C, respectively. Applying Ni/Ce-Zr as a catalytic filter in the sensor-filter combination results in selectivity to methane at 400ºC. This way the integration of the sensor and Au/Ce-Zr catalytic filter with dual selectivity has a higher performance than Ni/Ce-Zr filter.

Key words: Tin, Gas sensor, Gold, Nickel, Ceria-Zirconia, Catalytic filter, Selectivity

Introduction

Ethanol sensors have widespread application in wine making, breathalysers, medical processes and food industries [1]. In addition, methane as one of the pollutant and the major constituent of natural gas for domestic and industrial applications is necessary to be detected [2]. SnO_2 n-type semiconductor is the most favorable sensing material with high sensitivity for fabrication of chemical gas sensors utilized for detection of various VOCs. One of the methods for the selectivity improvement of a sensor toward a target gas is applying a catalytic filter. Gold nanoparticles and metal oxide supported gold nanoparticles are very active to oxidize ethanol, carbon monoxide, and several VOCs to CO_2 [3]. Moreover, Ni is one of the cheaper and highly active metal in most of the catalytic reactions and is better than noble metals economically [4]. On the other hand, the redox properties of CeO_2 could be enhanced by introducing additional nickel CeO_2 lattice via forming solid solutions.

Among the supports, ceria with high oxygen storage capacity (OSC) could be an appealing option and incorporation of zirconia in its lattice leads to higher redox property [5]. In this study, a dual selective sensor to ethanol and methane in the presence of CO as an interfering gas, using Au- and Ni-promoted Ce-Zr catalytic filter is introduced. Selectivity to methane by Ni/Ce-Zr catalytic filter is another achievement of the sensor-filter system.

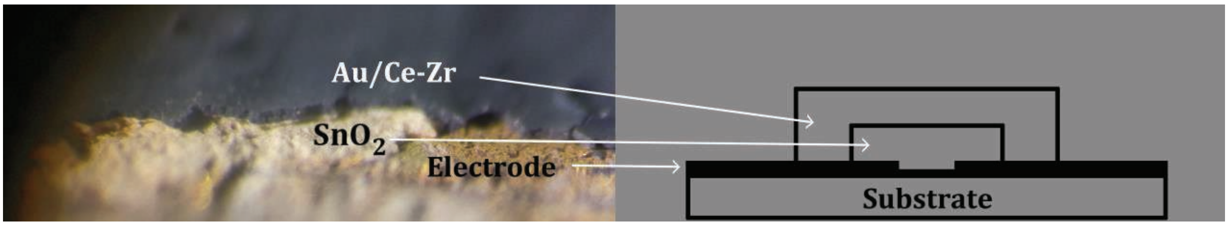

Fig.1. Optical microscope image of Au/Ce-Zr catalytic filter 70µm thick on the surface of the thick-film SnO_2 sensing material.

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Experimental

$Ce_{0.8}Zr_{0.2}O_2$ mixed oxide was prepared via a co-precipitation method, and then promoted with gold nanoparticles (1.0 wt. % Au/Ce-Zr) by a deposition-precipitation method. The ceria-zirconia was promoted with nickel by a dry impregnation method to 5.0 wt. % Ni/Ce-Zr. The SnO_2 fine nanoparticles were synthesized by a precipitation method. A flow system was used to measure the sensor response to 300 ppm carbon monoxide and ethanol and 5000 ppm methane at 100-480ºC in absence or presence of the filters.

Results, Discussions and conclusions

Fig. 1 shows an optical microscope image of Au/Ce-Zr catalytic filter with the thickness of about 70µm on the surface of the thick-film SnO_2 sensing material. Fig. 2a shows that Ni/Ce-Zr catalytic filter completely oxidizes CO and ethanol at 350 and 410ºC, respectively. The corresponding temperatures for Au/Ce-Zr (see Fig. 2b) are 220 and 310ºC, respectively. Figure 3 illustrates the sensor responses to carbon monoxide, ethanol, and methane in the presence and absence of the catalytic filter located on SnO_2 sensing layer. SnO_2 shows comparable responses to carbon monoxide and methane and much higher ones to ethanol. Using Au/Ce-Zr catalytic filter, dual selectivities to methane and ethanol are observed at 350 and 200ºC, respectively. While only methane selectivity at 400ºC is observed by applying Ni/Ce-Zr. filter.

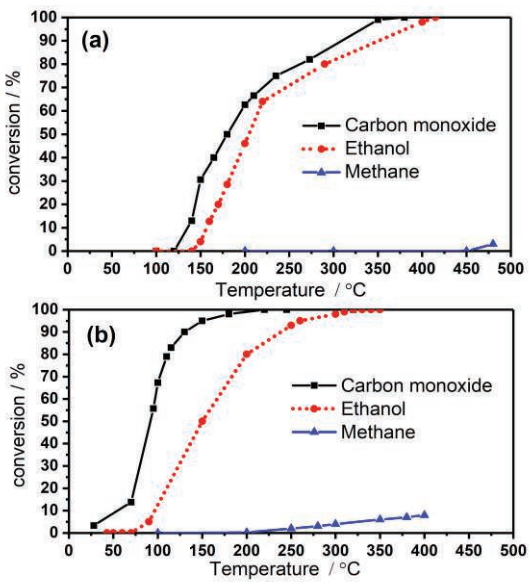

Fig. 2. The conversion-temperature curve of 300 ppm ethanol and carbon monoxide and 5000 ppm methane in air on (a) Ni/Ce-Zr and (b) Au/Ce-Zr catalytic filters.

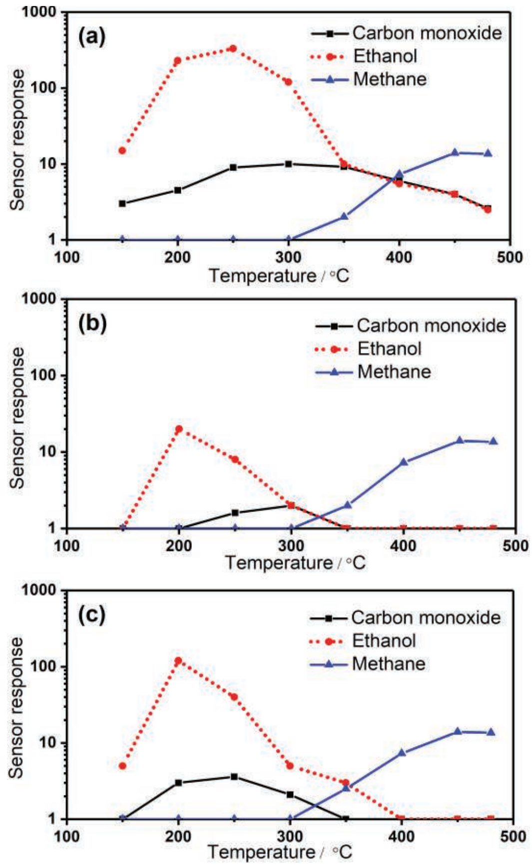

Fig. 3. Sensor responses to 300 ppm ethanol, 300 ppm carbon monoxide and 5000 ppm methane in air (a) without the filter (b) with Au/Ce-Zr catalytic filter (c) with Ni/Ce-Zr catalytic filter.

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