Solid State Science and Technology, Vol. 12, No. 1 (2004) 219-223

THE CHARACTERIZATION OF A SnO_2–CuO COMPOSITE–TYPE GAS SENSOR HAVING SENSITIVITY FOR H_2 GAS

Tulus Ikhsan Nasution, Ramli Omar, Ibrahim Abu Talib, Syahrial and Elvaswer

School of Applied Physics,
Faculty of Science and Technology,
Universiti Kebangsaan Malaysia,
43600 UKM Bangi, Selangor Darul Ehsan, Malaysia

Email : bunga_irwana@yahoo.com

ABSTRACT

The hydrogen gas sensing properties and the electrical conductivity of SnO_2-CuO composites were investigated at the temperature range of 125 - 400 ^oC in air and in 200 ppm H_2 gas. Four composite samples of SnO_2:CuO with the ratio of 4:1, 3:2, 2:3 and 1:4, respectively, were fabricated in the pellet form by mechanically pressing and sintered at 800 ^oC for 3 h in air. The electrical conductivity of the composites was found to increase with the increasing content of CuO and the SnO_2:4CuO composite showed the highest conductivity value. On the other hand, $2SnO_2$:3CuO composite was found to have the highest sensitivity to 200 ppm H_2 gas and may be suitable for use as a H_2 gas sensor.

INTRODUCTION

The single-oxide semiconductors such as SnO_2 and ZnO are widely used as sensing materials. Gas sensors made from these materials have been extensively investigated for detecting reducing gases such as H_2 or CO because they offer a simple sensing mechanism [1,2]. Sensing mechanism of the sensors are based on the changes in electrical resisitivity of sensors induced by the absorption and reaction of gas molecules on their surface. However, the main problems of such semiconducting oxides are insufficient gas selectivity, inability to detect very low gas concentrations, and changes in sensing properties caused by surface contamination [3].

In order to overcome the sensing problems inherent in the single-oxide semiconductor gas sensors, sensors consisting of mixtures of two or more oxides, which usually reffered to as a composite-type sensor, were proposed and extensively investigated [4]. For example, ZnO-SnO_2 composites which content more ZnO in their composition range were found to be more sensitive to CO gas at all temperatures than pure ZnO [5]. The 25% SnO_2 : 75% ZnO composite was reported to have high sensitivity to ethanol vapour [6].

In this study, SnO_2-CuO composite sensors were fabricated in a pellet form of various compositions. Current-voltage (I-V) characteristics of the sensors are tested in air and in 200 ppm H_2 gas. The effect of composition on sensitivity and electrical conductivity of the sample will be examined and discussed.

MATERIALS AND METHODS

SnO_2-CuO composite samples with different composition were prepared by using high purity commercial SnO_2 and CuO powders (Aldrich, 99.9 %). Powders were mixed in the molar ratios of SnO_2 to CuO equal to 4:1, 3:2, 2:3 and 1:4 in order to get four type of composite samples. The mixed powders were ground and then pressed into pellets and subsequently they were sintered at 800 ^oC in air for 3 h. The sintered pellets have approximately 12.95 mm in diameter and 1.75 mm in thickness.

For measurement of electrical properties, both side of flat surfaces of the sample was clamped by two plates of the sample holder. Current-voltage (I-V) characteristics of the samples were examined in air and in 200 ppm H_2 gas by using a potential method. The applied voltage was varied from –10 to 10 V and the temperature was varied from 125 ^oC to 400 ^oC. Current values were measured sequentially after equilibrating the samples for 20 min in air and 15 min in H_2 gas. Gas sensitivity was determined by the ratio of I_{gas}/I_{air}, where I_{gas} and I_{air} denote the current values in reducing gas and in air, respectively.

RESULTS AND DISCUSSIONS

The I-V plots for all the composite samples investigated in this study are nearly linear in the range of the tested temperatures, showing ohmic behaviour. For example, Figure 1 shows the I-V plots of $2SnO_2$:3CuO sample at 225 ^oC and 325 ^oC either in air and in 200 ppm H_2 gas.

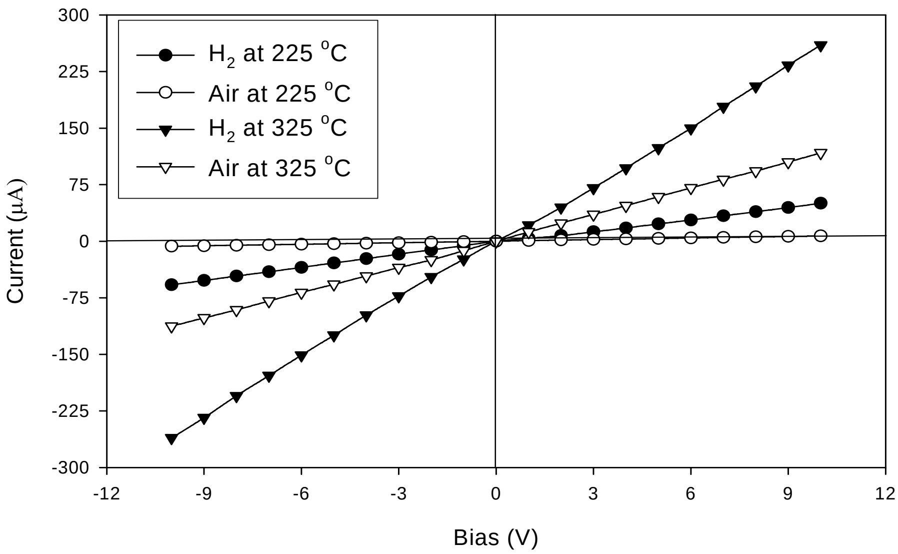

Fig. 1. I-V characteristics of $2SnO_2$:3CuO at 225 ^oC and 325 ^oC in air and 200 ppm H_2, respectively.

As can be seen that increase in current was higher when the sample was being exposed to H_2 gas compared with that in air, and also it increased with the increasing temperature either in air or in 200 ppm H_2 gas.

Figure 2. shows the electrical conductivity of the single-oxide SnO_2 and composite samples as a function of temperature ranging from 125 ^oC to 400 ^oC in air and in 200 ppm H_2 gas. Among all the samples, SnO_2:4CuO has the highest electrical conductivity. However, this sample shows no significant change of conductivity value upon exposure to H_2 gas as shown by the nearly overlapped curves with that in the air. While the other samples, eventhough they have lower electrical conductivity but they show a significant change of conductivity values upon exposure to H_2 gas. It is also shown that the conductivity of all composites are inclined to increase with the increasing content of CuO. However for SnO_2, above 350 ^oC, its conductivity decreases both in air and in 200 ppm H_2 gas. This could be due to the change of adsorbed gas species with temperature as was reported in [7].

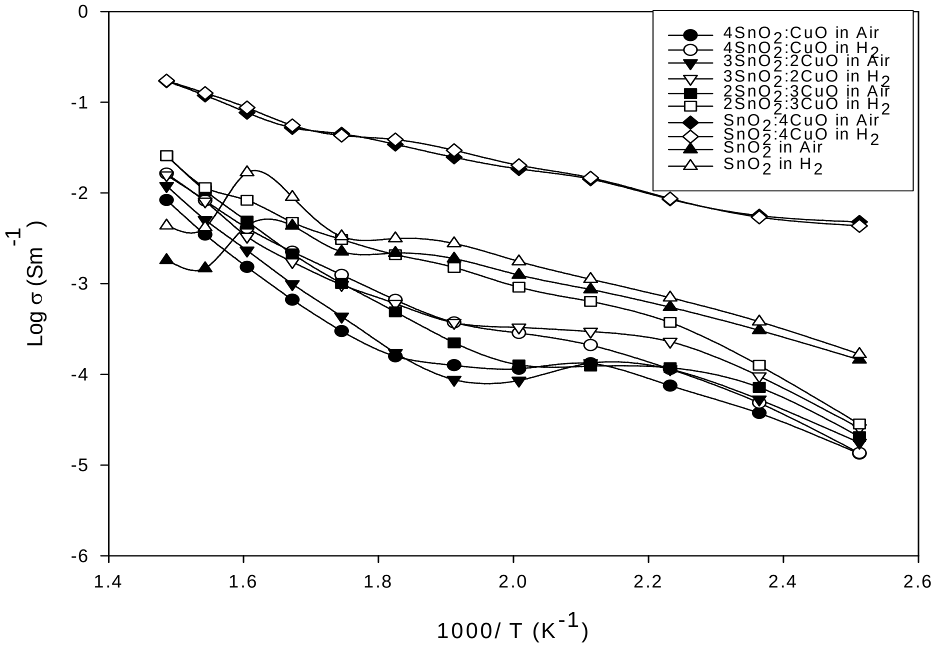

Fig. 2. Temperature dependence of electrical conductivity of the single oxide SnO_2 and SnO_2-CuO composites in air and 200 ppm H_2.

Figure 3. shows the sensitivities of the composite samples measured after 15 min exposure to 200 ppm H_2 gas as a function of temperature. The $2SnO_2$:3CuO, $3SnO_2$:2CuO and $4SnO_2$: CuO composite samples have the maximum sensitivity of ~9 at 225 ^oC, ~4.8 at 250 ^oC and ~4.6 at 275 ^oC, respectively. The sensitivity of SnO_2 sample is also shown in Figure 3 for comparison which has maximum value of ~3.9 at 350 ^oC. Thus, the $2SnO_2$:3CuO has the highest maximum value of sensitivity compared with that of the others. This behaviour may be associated with the amount of the formation of p-n contacts in the $2SnO_2$:3CuO which could be more compared with the other composite samples. On the other hand, no significant difference of the sensitivity values of SnO_2:4CuO upon exposure to 200 ppm H_2 gas with increasing temperature and thus was negligible. It is also shown that the addition of CuO up to 60 mol % lowered the temperature of the maximum H_2 gas sensitivity and increased the sensitivity to H_2 gas. This is expected to be due to CuO acted as a catalyst.

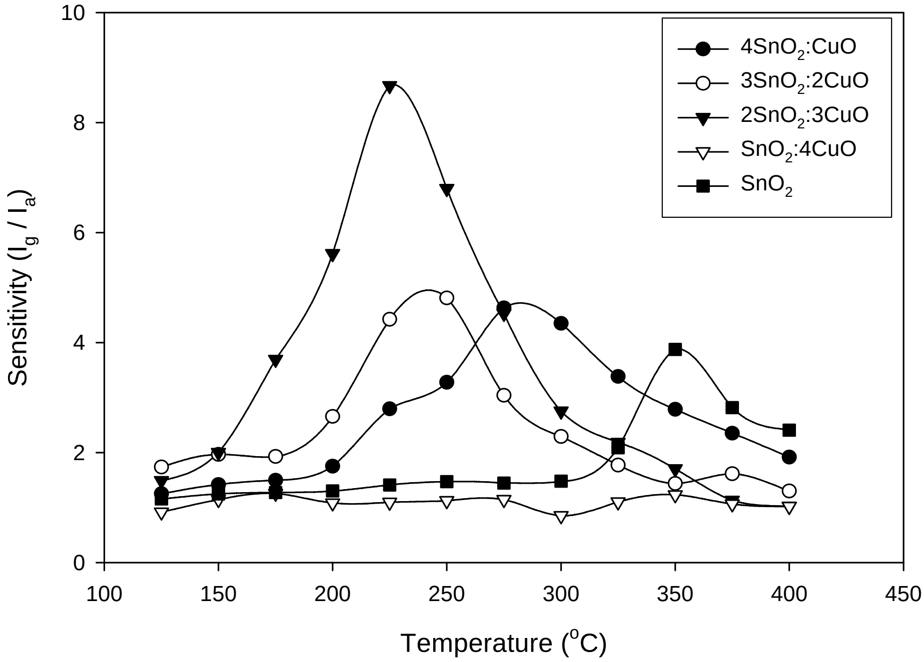

Fig. 3. Temperature dependence of H_2 sensitivity (200 ppm) of SnO_2-CuO composites.

Although in this study only a low concentration of H_2 gas (200 ppm) was used, but most of the composites, particularly the $2SnO_2$:3CuO sample, showed a significantly good response to H_2. Consequently, this composite sample may be suitable for use as a H_2 gas sensor.

CONCLUSIONS

The electrical conductivity and H_2 gas-sensing characteristics of SnO_2-CuO composites were studied. The conductivity of all composites was found to increase with temperature and the increasing content of CuO. The SnO_2:4CuO composite was observed to have the highest value of conductivity among all samples. Addition of CuO up to 60 mol% lowered the temperature showing the maximum H_2 gas sensitivity and increased the sensitivity of the sensors to H_2 gas. The $2SnO_2$:3CuO composite showed the highest H_2 gas sensitivity than that of the other composites.

ACKNOWLEDGEMENTS

IRPA grant (code:09–02–02–0039EA135) from MOSTE and the assistance of Mr. Mohd Nizam Mohamed and Mrs. Rokiah Mohd Yassin are gratefully acknowledged.

REFERENCES

[1] Jones, A., Jones, T. A., Mann, B & Firth, J. G. 1984. The effect of physical form of the oxide on the conductivity changes produce by CH_4, CO and H_2O on ZnO. *Sensors and Actuators. **5***. 75-88.

[2] Hauptmann, P. 1991. Sensors; Principles and Applications. 115-124. Prentice Hall. Salisbury. UK.

[3] Traversa, E. 1996. Beyond the limits of chemical sensors: novel sensing mechanisms for ceramics. In Progress in Ceramic Basic Science: Challenge Toward the 21 st Century. In Hirai, T., Hirano, S. T & Takeda, Y (Edt). The Basic Science Division. The Ceramic Society of Japan. 145-158. Tokyo.

[4] Yoon, D. H., Yu, J. H & Choi, G. M. 1998. CO gas sensing properties of ZnO-CuO composite. *Sensors and Actuators B.* **46**. 15-23.

[5] Yu, J. H & Choi, G. M. 1998. Electrical and CO gas sensing properties of ZnO-SnO_2 composite. *Sensors and Actuators B.* **52**. 251-256.

[6] Zhou, X., Cao, Q., Hu, Y., Gao, J & Xu, Y. 2001. Sensing behavior and mechanism of La_2CuO_4-SnO_2 gas sensors. *Sensors and Actuators B.* **77**. 443-446.

[7] Moon, W. J., Yu, J. H & Choi, G. M. 2002. The CO and H_2 gas selectivity of CuO-doped SnO_2-ZnO composite gas sensor. *Sensors and Actuators B.* **87**. 464-470.