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Synthesis and Characterization of Tin Oxide-MultiWalled Carbon Nanotube Composite Material as Carbon Monoxide Gas Sensor

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Abstract. Gas sensor based on nano-structured tin oxide (SnO_2) and multi-walled carbon nanotube (MWCNT) composite material has successfully been synthesized. Reflux method was used to produce SnO_2-MWCNT powder with various ratio 1:0, 4:1, 2:1, 1:1 and 0:1. The XRD result shows that the synthesized material comprises of the combination of carbon elements (MWCNT) and SnO_2 of the crystalite cassiterite phase. BET analysis shows that MWCNT particles increase the specific surface area of SnO_2 particles. SEM images show the morphology of the SnO_2 nanoparticle composite attached to the MWCNT wall with a diameter of 40-60 nm and dispersed around it. Gas sensor testing was carried out at room temperature, 50, 100, 150, 200, 250, 300, and 350^oC. All sensor samples were exposed to 30 ppm CO gas for 15 minutes. . It was found that sample with 1:0 ratio gives the best response with 98.91% at 350^oC. CO gas tests have also been carried out at various concentration 10, 30, 50 and 70 ppm to 4:1 SnO_2-MWCNT sample at 150^oC. The higher the CO gas concentration, the greater the response. SnO_2-MWCNT with 4:1 ratio at 50^oC has the fastest response time of 10 s and the fastest recovery time of 3 s.

Introduction

Air pollution is one of major problem in this world recently. One type of gas pollution which very toxic and threat human health is Carbon Monoxide (CO). CO gas is harmful to human health as it affects the human body's ability to bind oxygen [1]–[3]. CO gas directly binds to hemoglobin in the blood to form carboxyhemoglobin (COHb) instead of oxygen (O_2) [1]–[3]. The metabolism of cells, tissues, and human organs requires oxygen for the respiratory process. If COHb levels in the blood of more than 40% will result in respiratory failure and death [4-6].

Research in finding high performance of gas sensor continues to grow. The most popular gas sensor is metal oxide semiconductor [7]–[9]. Metal oxide semiconductor type materials have been widely used as gas sensors and have been tested on various types of gases. Basically, metal oxide semiconductor-based gas sensor material (SMO) must have a grain structure on nanometer scale that has a high surface area to volume ratio [8]. In addition, a good gas sensor must have high sensitivity, stable or resistant to heat and chemical treatment (high endurance), low power consumption (operating at room temperature), short response time and recovery time, and selective [10]. To improve sensor response and sensitivity, various precious metals such as Au, Pt, Pd and other nanoparticles are added to the SMO surface [11]–[13]. However, precious metals require high costs.

One SMO that has a good sensitivity to gas, when compared with other oxide semiconductors, is tin oxide (SnO_2). SnO_2 is a n-type semiconductor that has a wide band gap (3.6 eV at 300 K) so it can be utilized in various technologies such as lithium batteries, solar cells, catalysts, transistors and especially as gas sensors [3]. The working principle of SnO2 as a gas sensor is a change in electrical conductivity resulting from the reaction between oxygen ions and surface reducing gas. In general, SnO_2 nanoparticles that have large surface area make better sensitivity. This can be done by minimizing particle size [9]. In addition to controlling the morphology of SMO, another method to increase the performance of gas sensors is by combining with other materials such as carbon nanotubes (CNTs). The advantage of CNTs compared to other materials in terms of gas sensors is to have a fast response, low power usage and can operate at room temperature [14]. Applications of CNT-based gas sensors for the detection of methane, oxygen, and carbon dioxide have also been reported [15]–[18]. Other advantages are low chemical reactivity, has a low dimension that causes most of the atoms exposed to the environment and in other words has a large surface area, this causes this material to have a good sensitivity as a gas sensor [14]. Several studies have reported that SnO_2-MWCNT composites perform well in detecting NH_3, ethanol, methanol, H_2S, NO_2 and H_2 at room temperature [22-24]. Therefore, in this study we studied the effect of SnO_2-MWCNT composition ratio on carbon monoxide gas sensor performance.

Experimental

In this process, 0.5 grams of MWCNT is dispersed into 50 mL HNO_3, then ultrasonicated to separate the attached MWCNT-MWCNT so that the dispersion increases. The ultrasonication process was done for 30 minutes. The next process is to reflux MWCNT in nitric acid at 197^{o}C for 2 hours. In this process, assisted by heat, nitric acid will oxidize MWCNT walls and cut MWCNT. After 12 hours, the product was washed and rinsed several times by water and ethanol. MWCNT was then dried at 100 ° C for 6 hours.

0.5 M tin chloride dehydrate (4,513 gram) was dissolved into 40 mL ethylene glycol and then 5 drops of 3.5 M NaOH to make the pH 3. The solution was then refluxed for 3 hours at 190 ° C until the solution change color from clear to orange and white precipitate formed. Then the precipitate is filtered using centrifugal tubes and . After that the precipitate was rinsed with water and ethanol. The composites were then dried in an oven to evaporate the water and ethanol. Calcination was carried out at 500^{o}C for 2 hours to remove unwanted elements to form a pure SnO_2 powder.

To test the gas sensor, the material is deposited on an alumina (Al_2O_3) substrate that has been coated with a silver electrode. The distance between the electrodes is 6 mm. The method used in deposition is doctor blade. At first the composite powder is made into a paste by dissolving into ethylene glycol and arranged so that the formed pastes are not too thick and not too dilute. After the sensor material is deposited, it is then heated to 200^{o}C for two hours.

The performance of the gas sensor was tested by varying the temperature of the room temperature test; 50, 100, 150, 200, 250, 300, and 350^{o}C. The concentration of CO gas exposed was also varied at 10 ppm, 30 ppm, 50 ppm, and 70 ppm in the best sample and working temperature. Gas sensor testing scheme can be seen in Figure 1.

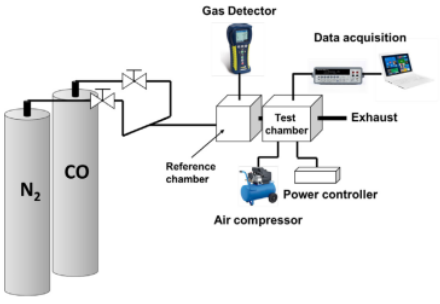

Fig. 1. Schematic of gas sensor measurement system

Result and Discussion

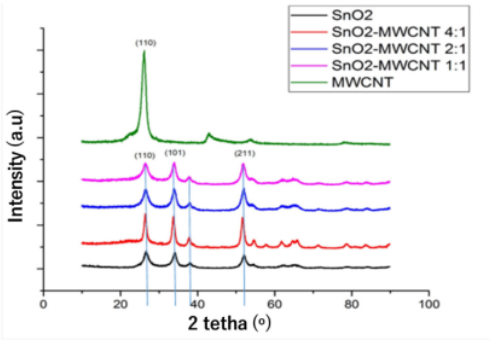

Fig. 2. X-ray diffraction pattern of SnO_2-MWCNT sample with ratio of 0:1, 4:1, 2:1, 1:1 dan 1:0

Figure 2 shows the diffraction pattern on 5 samples of SnO_2-MWCNT. The XRD results of SnO_2 show the peaks 110, 101, 200, 220, 211, 301 and 321 with diffraction angles at $2\theta = 26.7^o$, 33.97^o, 38^o, 51.8^o, 54.8^o, 66.1^o which is a cassiterite crystal phase with a tetragonal rutile structure (JCPDS No. 41-1445). The pure MWCNT XRD results show peak (110) at an angle of 26.14^o which is similar with graphite. In the 1:1, 2:1, and 4:1 diffraction peaks only seen in the crystalline phase SnO_2. The more MWCNT composition, the peak intensity of the composite sample is smaller.

From the Brunauer-Emmett-Teller (BET) we obtained the value of specific surface area of the material under test. The surface area of the SnO_2-MWCNT nanocomposite for each ratio of 1:0, 4:1, 2:1, 1:1, and 0:1 is 24.53, 29.66, 54.75, 82.67 and 153.22 m^2/g. The result of BET analysis shows the tendency that the more MWCNT mol composition in the sample, the more surface area we get.

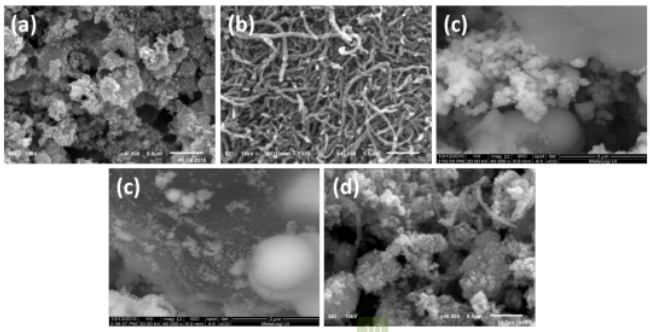

Fig. 3. SEM images of SnO_2-MWCNT with ratio of (a) 1:0, (b) 0:1, (c) 4:1, (d) 2:1, (e) 1:1

The morpholgy of all samples were analyzed using Field Emission Scanning Electron Microscope (FESEM) as shown in Figure 3. Figure 3 shows sample morphology for MWCNT, SnO_2, and SnO_2-MWCNT composites with ratio of 1:1, 2:1, and 4:1. From the micrograph shown in Fig. 3 (c), (d), and (e), the MWCNT tube has been coated by the white particles on the surface. The white particles are believed to be the SnO_2 nanoparticles that have been attached to the surface of the MWCNT tube. MWCNT has an outside diameter varying between 30 - 60 nm. MWCNT formed randomly resulting in many junctions between the tubes formed. The MWCNT wall is successfully decorated by SnO_2 and 2 µm size SnO_2 partially agglomerated. It is also seen the effect of differences in the ratio of composition to the material formed. The higher the concentration of SnO_2, the less visible MWCNT is. However, good dispersion is seen in the 2:1 composition. The particle size of SnO_2 in each composition is in the range of 50 nm-2 µm. The presence of MWCNT has no major effect, although with increasing agglomeration of SnO_2, the agglomeration is not perfect, only particles attached to each other was seen.

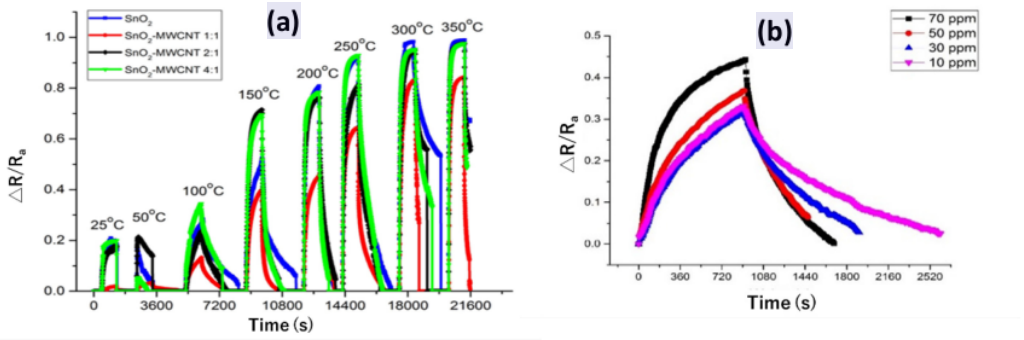

Fig. 4. (a) SnO_2-MWCNT response exposed to 30 ppm CO gas and (b) Response of 4:1 sample at 150^{O}C

Figure 4 (a) shows SnO_2-MWCNT sample responses that exposed to 30 ppm CO at various working temperatures from room temperature to 350°C. From 100 ° C to 350 ° C, in all sample it exhibited response with a decrease in the surface resistance after exposure to 30 ppm CO gas for 15 minutes. This result indicate that all composites exhibite n-type semiconductors. Furthermore, this result also shows that in that temperature range, SnO_2 acts as an active sensing material in SnO_2-MWCNT composite. While at room temperature and 50^{o}C in all samples there is an increase in resistance when exposed to CO gas. This phenomenon is a physisorption mechanism (Van der Waals interaction) at low temperatures because the energy involved in this mechanism is relatively low so it can occur at low temperatures, but the resulting response is low [18].

Table 1. Sensitivity of SnO_2-MWCNT againts 30 ppm CO gas

Temperature (^{o}C)	Response SnO_2:MWCNT (%)				
	1:0	4:1	2:1	1:1	0:1
25	25.81	19.99	17.83	1.97	0.63
50	17.14	5.46	21.45	3.37	0.26
100	28.36	34.48	23.79	13.29	0.77
150	52.47	69.52	71.5	39.92	0.66
200	80.81	78.44	76.31	48.69	0.9
250	92.28	92.94	81.5	64.63	1.49
300	98.29	95.31	94.97	83.57	1.47
350	98.91	97.62	97.42	84.28	-

MWCNT worked better at 100 and 150^{o}C. Moreover, in the sample with the ratio of 2:1 and 4:1 at 100 and 150^{o}C temperatures the responses are better than the pure SnO_2 sample at the same temperature. For working temperatures above 200^{o}C, a pure SnO_2 sample (1:0) seems better in response to a 30 ppm CO gas. The best response was recorded in the SnO_2 sample at 350^{o}C working temperature with a value of 98.91%. The more MWCNT composition, the lower the response of sensors at high temperatures. This is due to the MWCNT particles inhibiting the active site of SnO_2 which is more sensitive to exposure to CO gas on the surface. Sensitivity value can be seen in Table 1.

Figure 4 (b) shows the difference of response value at four variation of CO concentration. The higher the CO gas concentration, the better the response. As metal oxide semiconductor gas sensor theory stated, at certain temperature, n type semiconductor will adsorbed oxygen gas [18]. The gas then dissociated and ionized by taking electron from the surface oxide lead to increase the oxide's resistance. When reducing gas such as CO introduced to the surface, CO reacted with the oxygen ion (O_2^-, O^-, or O^{2-}) resulted in CO_2 as a product, released the electron back to the surface and decrease the resistance [18]. The increasing response with increasing the CO concentration is due to the increasing the amount of reducing gas which can reacted with oxygen ion. Thus, it will affect the number of electrons released from the O^{2-} or O^-ions back to the sensitive surfaces of the SnO_2-MWCNT composite.

Summary

The SnO_2-MWCNT composite material was fabricated by combining MWCNT and SnO_2 crystal-phase cassiterite. MWCNT increases the specific surface area of SnO_2 particles. All SnO_2-MWCNT sample compositions have a CO 30 ppm gas response at room temperature. The highest response value was achieved by SnO_2-MWCNT 1:0 sample with 98.91% at 350 ^oC working temperature. However gas composite with ratio 4:1 show good response at 100^oC which is relatively low temperature for gas sensor application. The SnO_2-MWCNT 0:1 sample is not sensitive to CO 30 ppm gas with a sensitivity value of less than 1.5%.

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