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An Ohmic Contact Behavior of Flexible Thick Film Carbon-Based Electrode with TiO_2 Layer at Different Operating Temperature

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Abstract

An electrical property of a carbon-based referring to graphite and multiwall carbon nanotube (MWCNT) as conductive paste with titanium dioxide TiO_2 were presented in this research work. Screen printing technique has been selected to deposit graphite and MWCNT thick film respectively on kapton film substrate. Each conductive paste was fired at 150^0C before printing TiO_2 paste printing on top as metal-oxide layer followed by another firing at 350^0C. The electrical properties measurement was performed using Current-Voltage (I-V) characteristics measurement from -10 V to +10 V, at various temperatures from 28, 50, 100, 150, 200, to 250^0C. The I-V results have shown linear and symmetric I-V relationship indicating good Ohmic contact between carbon based and TiO_2 thick film. The resistance value of graphite has a low value compared to MWCNT at room temperature measurement. Meanwhile the resistance values for Graphite/TiO_2 and MWCNT/TiO_2 showed a decreasing trend again increase the temperature operations associated with slower electron mobility effect of TiO_2.

Keywords: Ohmic contact, Thick film, Graphite, Multiwall carbon nanotube, Resistance.

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1. Introduction

The term "Ohmic" refers in principle to a contact that does not inject and has a linear I-V characteristic in both directions. In practice, a contact is considered Ohmic if the voltage drops across it is significantly smaller than that across the device. Linearity of the I-V relationship is less important if the contact resistance is very small compared to the device resistance. Another important characteristic of Ohmic contacts in semiconductor devices is their reliability and repeatability. In particular, the contact material should not undergo electro migration under high electric fields or modify active structural characteristics during device operation. It is often required that the thermal impedance of the contact below remove heat from the device.

Recently, carbon based conductive paste has been developed to replace current conductive paste such as copper, silver and platinum which is not appropriate for flexible applications. Carbon based conductive paste in this study represent graphite and multiwall carbon nanotube (MWCNT) which behave as an allotrope of carbon. Graphite is carbon that can be produced as natural or synthetic graphite. Graphite has a planar structure containing carbon atoms in each layer. Meanwhile MWCNT is a special form of carbon nanotubes in which multi single walled carbon nanotubes are nested inside one another. Graphite and MWCNT conduct electricity due to vast electron delocalization within the carbon layers, the valence electrons are free to move and able to conduct electricity [1]. Carbon based conductor suitable as thick film at flexible substrate. The flexible electronics circuit is the most popular in the industry due to its diversity in applications and design. kapton polyimide film is used as a substrate in this research work because of its flexibility. Technical depositions are made by various methods such as photolithography, screen printing, inkjet printing, and other methods [2-3]. Carbon based interdigitated electrode is easy to be deposited by using flexible substrate; compared to alumina substrate since kapton substrate is flexible, more robust, and not fragile. Thick film carbon based via screen printing fabrication process is a simple, low-cost, and time-saving method. The electrode material adopted is also important as it will increase the sensitivity and reliability of the sensor. Metals are commonly used as electrodes, such as silver, gold, and platinum, due to their low resistivity [4]. Among these materials, the most popular material is silver because it is the cheapest and most stable in the air. However, silver tends to be easily oxidized under a high humidity environment [5]. The purpose of this research work is to identify the capability of graphite and MWCNT as the replacement material for electrodes choice in the sensor platform itself. Including comparing the I-V characteristics of two different conductive paste materials and thick film layer of TiO_2 prepared using the screen-printing technique.

2. Materials and methods

Graphite and MWCNT conductive paste as carbon-based interdigitated electrode components were provided from Serdang Paste Technology Sdn. Bhd. Inorganic compound of TiO_2 (Aeroxide P25) was purchased by Sigma-Aldrich and prepared as thick film. Powder TiO_2 are mixing with linseed oil binder as ratio of 70:30 to form the paste. In order to create an organic linseed oil binder, linseed oil (85 wt%) and m-xylene (12.5 wt%) are combined at 250 rpm for 3 hours at 40^0C. Then continued by mixing α-terpineol (2.5 wt%) using a magnetic stirrer for 250 rpm for 3 hours at same temperature 40^0C to obtain a homogeneous organic binder [6]. The advantages of linseed oil used in this research are particularly good adhesion to surface like metal, wood, and plastic. The screen-printing fabrication method was used in paste deposition to fabricate a thick film. **Figure 1a** shows the process to develop layer of interdigitated electrode and TiO_2. By using squeegee to force and push equilibrium the paste on the screen mesh with design 10.0 mm x 10.0 mm square was printed on the substrate. The thick film used in this research work consists of two film layers. The first layer is carbon based conductive paste, and the second layer is thick film TiO_2. The first layer was annealed at 150^0C for 30 minutes and rested before depositing the second layer on the top of it. The process continues for deposited TiO_2 paste at 350^0C for 60 minutes. The layer of interdigitated electrode on flexible kapton and alumina as shown in **figure 1b**. The size of the film is 4.25 x 4.25 mm, while the flexible substrate size was 21.45 x 25.20 mm. Ohmic contact behavior depends on electrical properties of Current-Voltage (I-V) characteristics. The I-V characteristics on sample were studied using a two-point probes measurement Keithly and conducted by LabView software. Copper wires were attached to both ends of the sample with thick film TiO_2 on top of it. A current (I) flow through the sample because of a voltage source from a Keithley 2400 Source Meter applying a range of voltages from -10 V to +10 V across it. The operating temperature was controlled from Autonics Temperature Control to supply a temperature operation beginning from 28, 50, 100, 150, 200, and 250^0C. Two-point probe measurement on carbon-base conductive paste and TiO_2 materials has been carried out in order to observe the resistance value at different temperature operations.

3. Results and Discussions

The morphological structure of graphite and MWCNT were annealed at a temperature of 350°C, as shown in **figure 2b** at magnification 50,000. **Figure 2c** shows the magnified images for the image at a magnification of 50,000 and 100,000 for graphite and MWCNT, respectively. The images revealed that the particle formed the typical microstructure of graphite, where starched layers of graphite are observed. Graphite was also observed coated around the binder because the temperature used was not sufficient to remove all the binder at a temperature of 350°C. Meanwhile, a small tube of MWCNT was also coated by the binder for the same reason. However, the binder for both pastes should be processed solely through the drying of elements in the binder. The resistance value of each carbon-based mentioned in **Table 1** was measured by using a two-point probe. The resistance of the sample is given by R equal to resistance in Ω, V is voltage in volts, and I is current in Amperes. The sample on alumina produced resistance value of graphite was 171.53 Ω and MWCNT 534.33 Ω respectively. Carbon-based on flexible substrate kapton decrease the resistance 342.95 Ω for graphite and 743.28 Ω for MWCNT. It shows graphite has low resistance value compared to MWCNT thick film. It is also revealed that graphite gives higher conductivity compared to MWCNT on alumina without TiO_2 layer. After depositing the thick film TiO_2 layer on the top of interdigitated electrode, the resistance value is increase for both substrates. Graphite/TiO_2 resistance on alumina 4.423 x 10^3 Ω compared to on Kapton substrate it increases to 11.804 x 10^3 Compared on substrate alumina and kapton for MWCNT/TiO_2 layer the resistance were increase from 8.546 x 10^3 to 36.130 x 10^3 Ω. Both situations show the slow of electron movement because of electron mobility slow on TiO_2 as resistivity movement. **Figure 3** shows the graph resistance (Ω) versus voltage (V) from -10 V to +10 V at difference operations temperature starting room temperature, 50, 100, 150, 200 and 250 ^{0}C. The observation from both substrates produces I-V characterizations graph that shown an Ohmic contact behavior.

It verifies a good conductivity produced from carbon-based graphite and MWCNT electrode with thick film layer of TiO_2. **Figure 3 (a) and (d)** shows carbon-based deposited on thick film TiO_2 layer on alumina substrate show the behavior of Ohmic contact. Meanwhile, **Figure 3 (b) and (e)** show behavior on polyimide kapton as flexible substrate. Both show the behavior of Ohmic. Increase the temperature from room temperature to 250^0C, show decreasing the resistance value. It is because the mechanism of current flow depends on temperature. As the temperature increases, some electrons of the semiconductor acquire energy and become free for conduction. The free charge particles that carry the charge collide with the atoms more frequently as they try to go through TiO_2. This phenomenon causes some extra resistance in the path of free electrons. Hence, resistance decreases with an increase in temperature. **Figure 3 (c) and (f)** show the illustrations of graphite/TiO_2 and MWCNT/TiO_2 where the voltage +10 Volt was supplied to the carbon-based layer and flow to the TiO_2 layer before coming back to the circuit. Electrons immigrate from TiO_2 layer before carbon-based layer harvests the electrons from the TiO_2 and close to the circuit. **Figure 4** presents the resistance (Kilo Ohm) versus Temperature (°C) shown straight line graph for graphite and MWCNT electrode with different temperature contents.

Graphite and MWCNT electrode on alumina substrate has gradients value -0.0230 and -0.0042, both shown the effect for temperature decrease the resistance value because of more electrons free in conductivity increase. Compared to gradient value for flexible substrate graphite and MWCNT electrode shown value -0.0110 and -0.0330 respectively. MWCNT electrode on kapton show a higher resistance value compared to another electrode. Without TiO_2 layer, MWCNT electrode on the flexible substrate kapton show the good linear graph and higher resistance compared to another electrode. The resistance values of graphite/TiO_2 and MWCNT/TiO_2 were measured using I-V testing at different temperatures between 28 to 250°C. The linear graph for both samples in **figure 5 (a) and 5 (b),** showing the MWCNT/TiO_2 than graphite/TiO_2. The value of gradient for MWCNT/TiO_2 was -0.0063 has shown a higher value compared to graphite/TiO_2 -0.00203. Each resistance was decreased when the temperature increased. The mobility of an electron is easier when the temperature increases due to free electron produced [7-11]. The results show the electrical conductivity increase while the resistivity decrease when annealing temperature become higher because of the recrystallization occurs during the annealing process. Graphite is an electrical conductor. It can conduct electricity due to the vast electron delocalization within the carbon layers. These valence electrons are free to move, so can become conductivity due to changing of temperature [12-14].

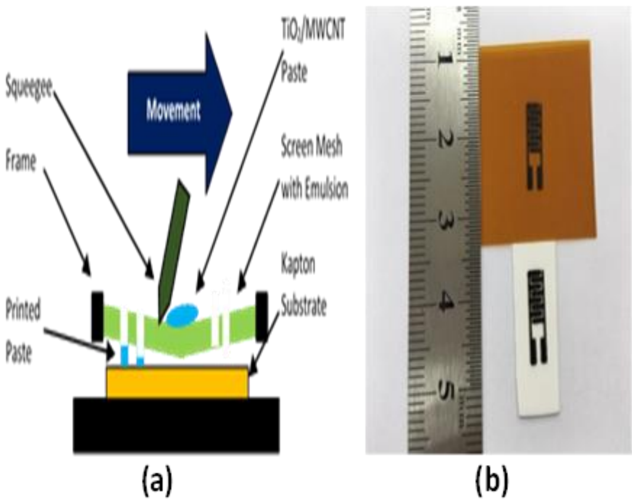

Figure 1: Illustration of depositions carbon-based electrode and TiO_2 thick film layer (a) screen printing technique apply to TiO_2 paste on screen mesh by squeegee on top of substrate and firing at 350 ^{0}C for 60 minutes (b) sample carbon-based electrode on the film dimensions 4.25 x 4.25 mm by using alumina, while the flexible substrate size was 21.45 x 25.20 mm on kapton substrate.

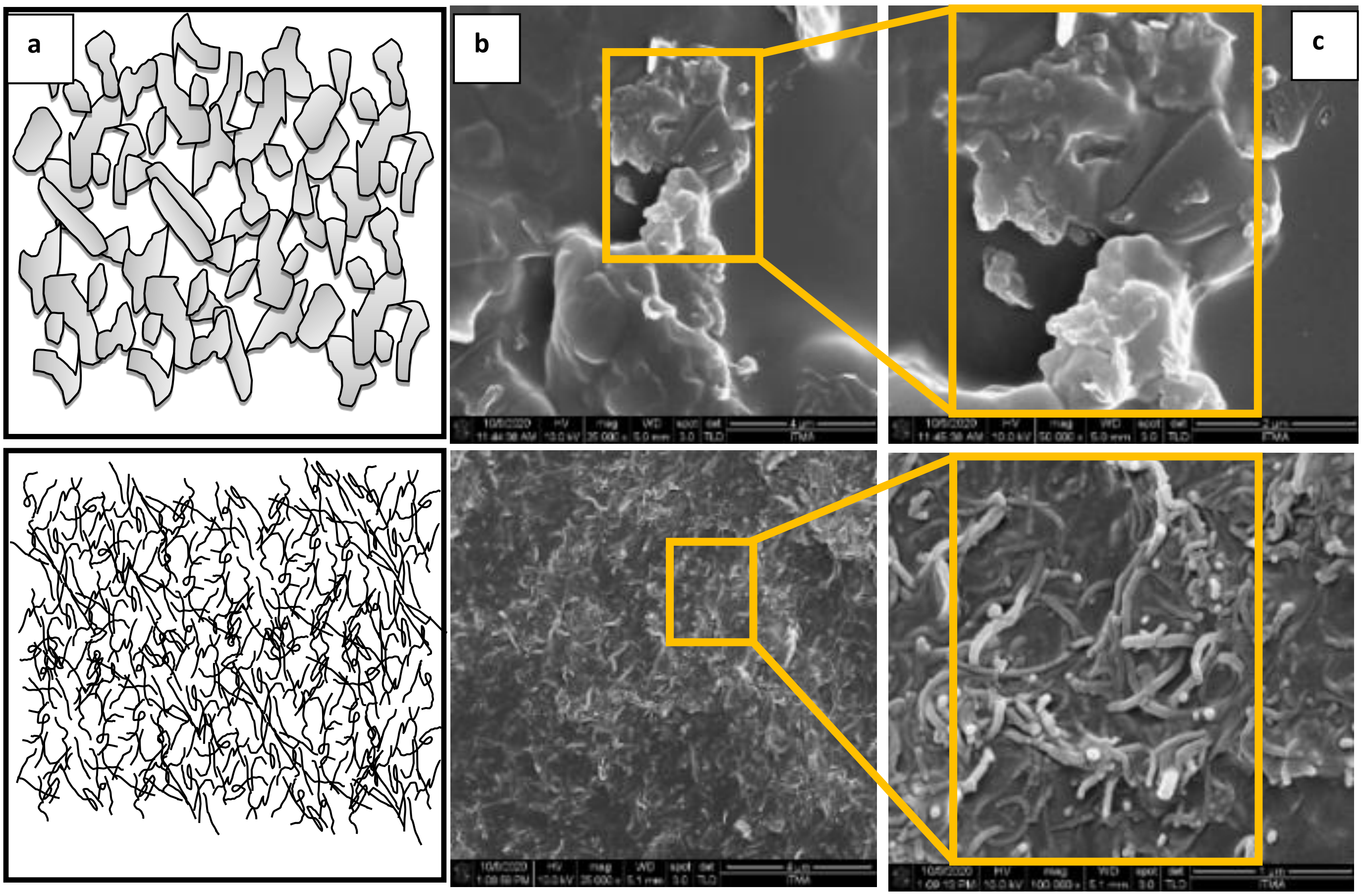

Figure 2: FESEM image morphology structure of graphite and MWCNT; (a) drawing schematic illustration; (b) magnification 25,000 and (c) magnification 50,000 and 100,000 respectively.

Table 1: Resistance value of graphite and MWCNT at room temperature.

Materials Layers	Type of Substrate	Resistance (Ω) without TiO_2	Resistance (Ω) withTiO_2
Graphite	Alumina	171.531	4.423 x 10^3
	Kapton	342.952	11.804 x 10^3
MWCNT	Alumina	534.333	8.546 x 10^3
	Kapton	743.284	36.130 x 10^3

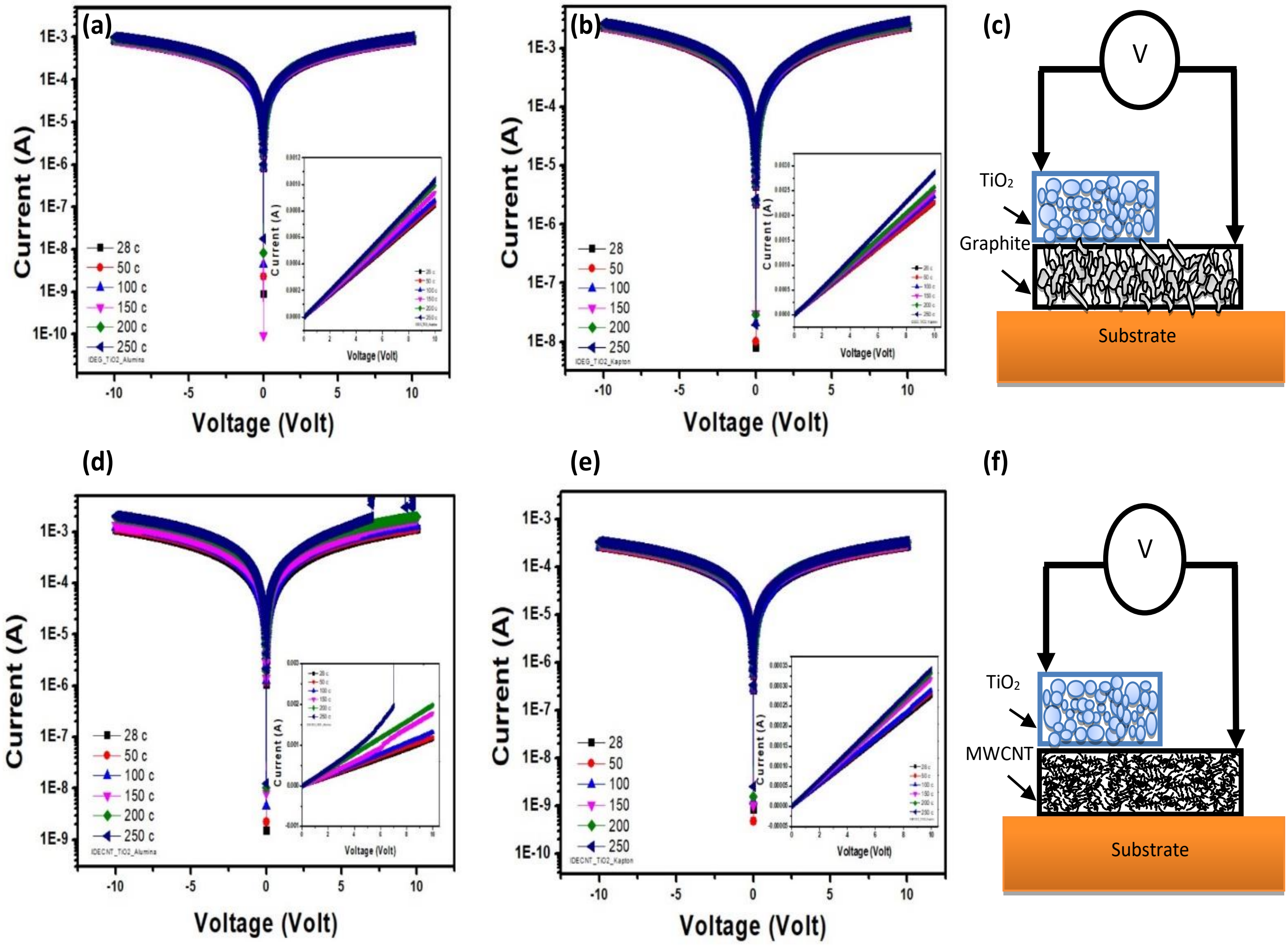

Figure 3: Current-Voltage (I-V) Characterizations on carbon-based electrode at difference operating temperature and difference substrate; (a), (d) sample on alumina substrate; (b), (e) sample on flexible substrate Kapton; (c), (f) illustrations of graphite/TiO_2 and MWCNT/TiO_2

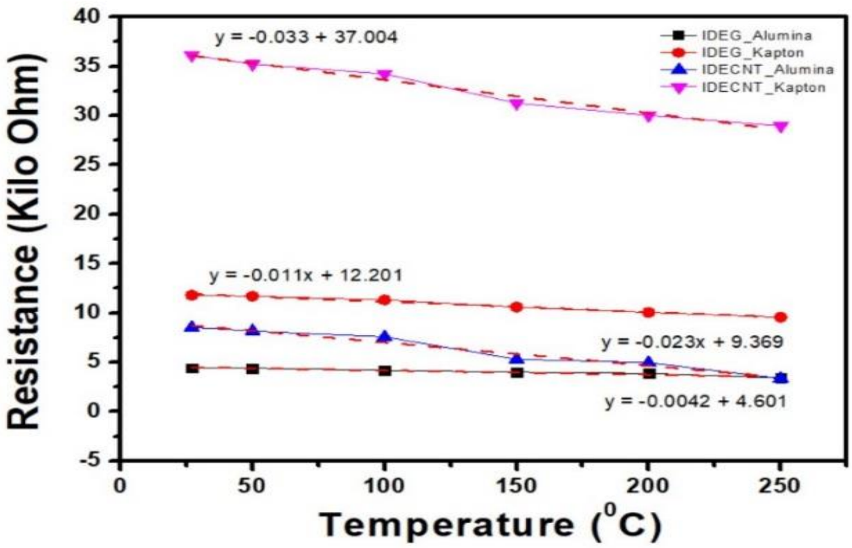

Figure 4: Resistance (Kilo Ohm) versus temperature (^{0}C), for electrode graphite and MWCNT on alumina and kapton substrate on different operating temperature.

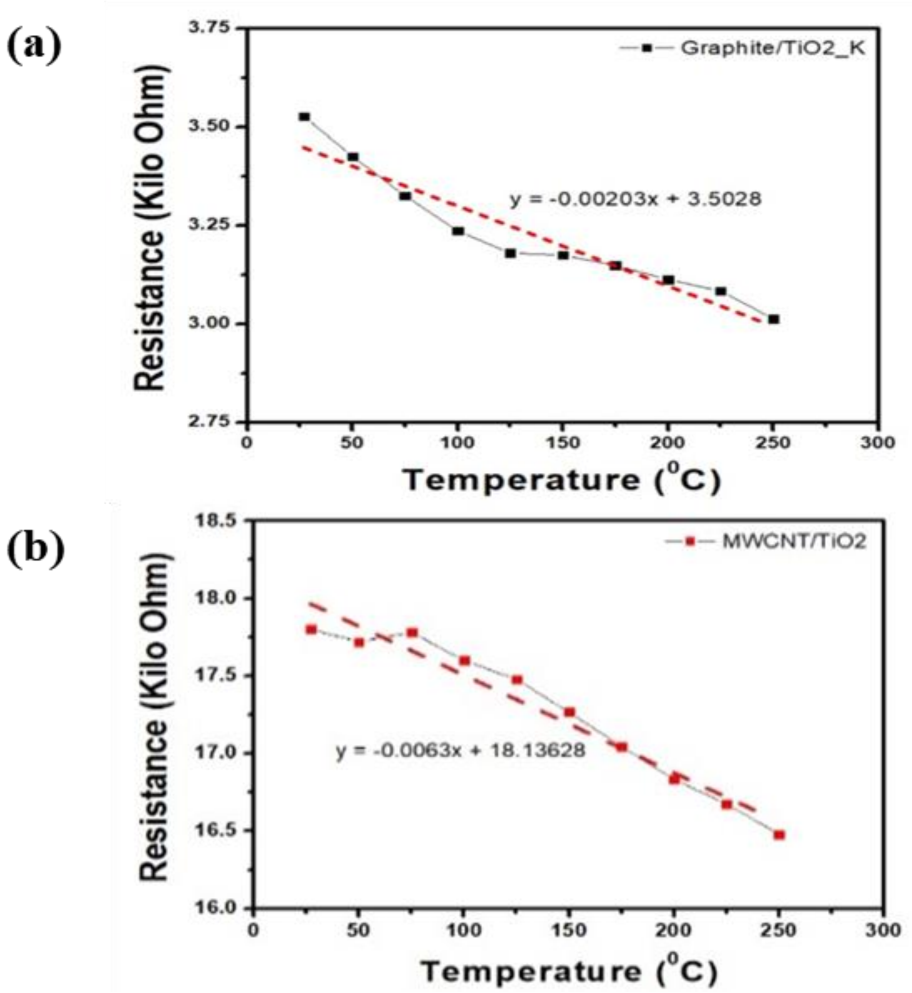

Figure 5: Resistance (Kilo Ohm) versus temperature (°C); on flexible substrate kapton for (a) graphite/TiO_2 and (b) MWCNT/TiO_2 at different operating temperature.

4. Conclusions

The effect of different materials via graphite/TiO_2 and MWCNT/TiO_2 with has been studied in detail in this research work. I-V characteristic measurements technique has been used to evaluate the current and resistance of the sample according to Ohmic contact behavior. The measurements have shown that the resistance value has a current flow through the sample and carries a good response in graphite/TiO_2 layer as compared to MWCNT/TiO_2. The performance of both samples exhibited an Ohmic contact behavior at high temperature decrease the resistance value. The TiO_2 sensing layer effect on graphite and MWCNT electrode causes an increase in the resistance value due to slower electron mobility by introducing a metal oxide. The carbon-based electrode to semiconductor sensing layer of TiO_2 as supported by experimental data. Based on this study the carbon-based is more suitable to replace current conductive material as used as flexible electronic applications such as gas sensor, pressure sensor and microwave sensor.

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