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PREPARATION AND CHARACTERIZATION OF ZINC OXIDE ON GREEN CARBON NANOTUBE COTTON

J.M Yusof[1], I. Ismail[1], Rahimi M Yusop[2], S.A Rashid[1], M. A. Mat Nong[1], R. Osman[3] and I.H Hasan[3]

[1]*Material Processing and Technology Laboratory, Institute of Advanced Technology, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia*

[2]*School of Chemical Sciences and Food Technology, Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Selangor, Malaysia*

[3]*Functional Devices Lab, Lab, Institute of Advanced Technology, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia*

**Corresponding author: juraina@upm.edu.my*

ABSTRACT

Recycling waste cooking palm oil (WCPO) as a source for alternative fuel such as biodiesel and valuable carbon materials such as carbon nanotubes (CNT) is a way to exploit it from waste to wealth. Carbon nanotubes cotton (CNTC) is one of the carbon nanostructures synthesized in high-temperature furnace. It resembles cotton candy, composed of low spatial density, ultra-long individual-al CNT, hydrophobic, oleophilic and exhibits good electrical and mechanical properties. CNTC from WCPO was synthesized using in-house chemical vapor deposition reactor. Ferrocene was used as catalyst with thiophene as a growth rate enhancer. The synthesis was carried out at temperature ranging from 1000 °C to 1200°C. The electrodeposition of ZnO was performed in a three electrode electrochemical cell with CNTC as the cathode. The result obtained showed that ZnO was successfully deposited on the CNTC. The combination of ZnO and CNTC hybrid is a promising composite that can be further explored in the application of piezoelectric nanogenerator, sensors and high-performance electromagnetic absorbing material.

Keywords: CNT cotton; waste cooking palm oil; zinc oxide

INTRODUCTION

Cooking palm oil is widely used in the consumer food chain and the demand has escalated over years as consumers consumptions becomes higher due to the influx of food and beverages manufacturing industry. Global oils and fats production is projected to expand by approximately 1.45% to 209 milliotonnes in 2017, compared to 206 million tonnes in 2016 [1]. Due to that, the amount of waste cooking oil has increased simultaneously which leads to uncontrolled dumping and discharging to the environment. This has triggered major concerns over its effects towards the environment in terms of ecosystem and biodiversity stability. Recycling waste cooking palm oil (WCPO) as a source for alternative fuel such as biodiesel and valuable nanostructures materials such as carbon nanotubes (CNT) is a way to exploit it for a better usage. The advantages of using WCPO as a carbon feedstock in carbon nanomaterial synthesis are because it is inexpensive, readily available and has no issue on shortage of supply. Carbon nanotubes cotton (CNTC) is one of carbon nanotubes superfibre materials that resembles cotton candy, composed ofvarious length of individual CNT with low spatial density, hydrophobic and oleophilic. In addition, it also exhibits good electrical and mechanical properties in terms of highly conductive, withgood mechanical strength. This makes it as a promising substrate with outstanding charge carrier mobility, which can be utilized to improve the performance of piezoelectric nanogenerator as compared to the common substrates such as silicon and metals.To date, most of the research attempts in Malaysia using WCPO mostly in the expense of turning it into inexpensive feedstock for biodiesel production. However, a research led by a local group demonstrated that WCPO from domestic frying can be utilized as an efficient, economical and environmentally friendly carbon source for the production of high purity CNTs [2]. Another research also reported on refined vegetable oil replaced by WCPO as a source of triglycerides for biodiesel production [3]. WCPO was further explored as a carbon source to produce high purity CNT around 87% and high crystallinity for supercapacitor application [4]. A novel flexible nanogenerator made of ZnO nanoparticles and multiwall carbon nanotubes mixed with polidymethylsiloxane (PDMS) resulted in high output voltage when bending, comparable to other materials as piezoelectric nanogenerator [5].

EXPERIMENTAL

CNTC from WCPO was synthesized using chemical vapor deposition (CVD) reactor. Ferrocene was used as catalyst with thiophene as growth rate enhancer. WCPO was refined physically to remove solid impurities then heated at 100°C above to eliminate excess moisture and any other products of low melting points from the frying process. Then it was mixed with ferrocene and thiophene with a weight ratio of 90.34%, 5.33%, and 4.33% respectively. Synthesis was carried out by heating the tube furnace for 30 minutes with 300sccm argon flow at 450°C to eliminate the amorphous carbon left in the tube. The tube temperature was ramped up to 1150°C and a flow of hydrogen at 150sccm was passed through the tube for 30 minutes. The mixed solution in a 40ml syringe was loaded into a syringe pump which was connected to the CVD. Meanwhile, electrochemical deposition of zinc oxide on CNTC was performed by a potentiostat VersaStat 3 with CNTC as the cathode, Pt as anode and saturated calomel as the reference electrode. The electrolyte was a solution of $ZnSO_4.7H_2O$ and $(NH_4)_2SO_4$ in distilled water. A piece of CNTC was cut into $2.25cm^2$ and clamped to the electrochemical cell as working electrode. The synthesis was carried out at room temperature for 500s at potential -6.8v. Afterwards, samples were washed with distilled

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water and dried. The structural and morphology studieswere carried out using Field Emission Scanning Electron Microscopy (FESEM).EDX analysis was performed to identify the elements presence in the sample.Raman spectrum of the sample was examined by DXR Raman Microscope (Thermo Scientific) with 25µm pinhole aperture, excitation wavelength of 532nm and 10 mV laser power. The chemical composition and crystallographic structure of the samples was examined by D8 AdvanceBruker AXS, X-Ray Diffractometer (XRD).

RESULTS AND DISCUSSION

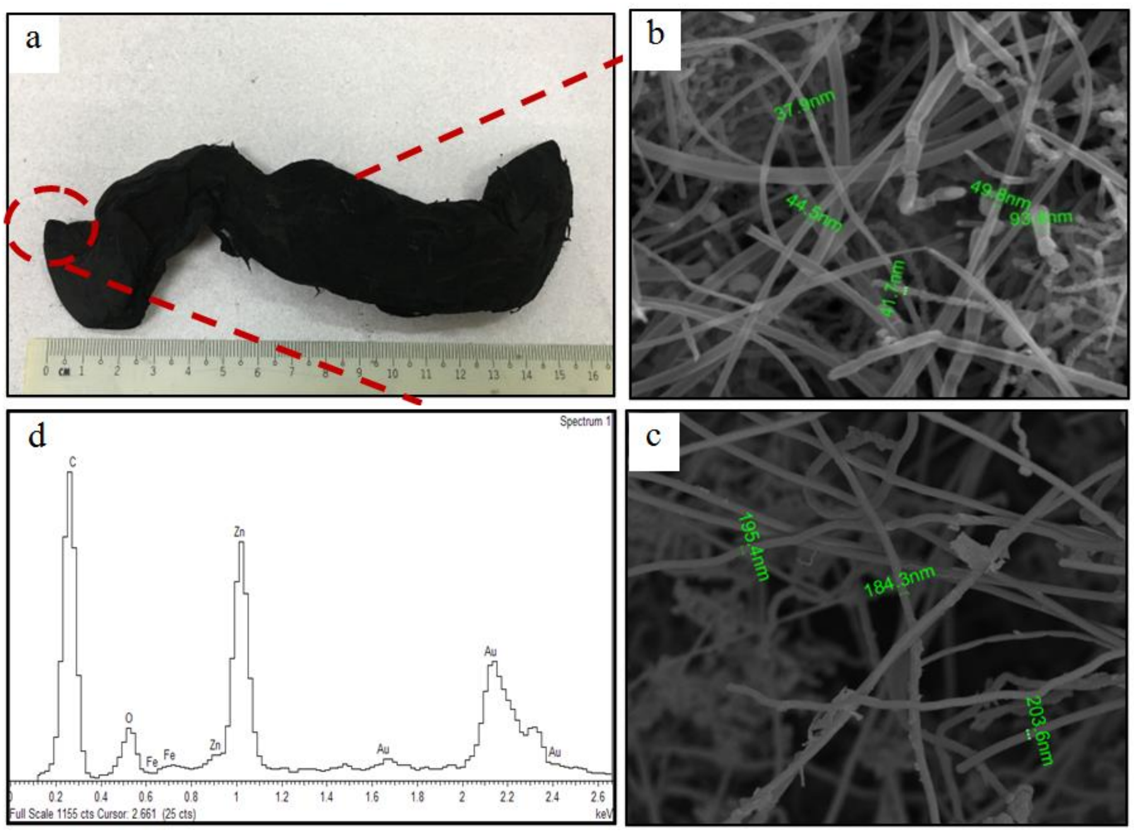

Figure 1:(a) CNT cotton , FESEM images of (b) CNT cotton, (c) CNTCZnO hybrid (d) EDX analysis of CNTCZnO

The physical appearance of carbon nanotubes cotton (CNTC) which resembles cotton fibre or cotton candy that is soft, fluffy and lighweight can be seen in Figure 1(a). Meanwhile, under FESEM high magnification image as seen in Figure 1(b) it was observed that the CNTC consisted of a mixture of long and short truncated nanotubes on the order of a few tens of micrometers. The variation of nanotube length is due to the concentration fluctuation of carbon source flows in the furnace. The presence of graphitic foliates along some the tubes length which resembles graphenated carbon was also observed from the image. Figure 1(c) shows the image of CNTC after being hybridized with ZnO. It can be observed that the CNTs diameter was getting bigger due to the coatings of ZnO along its tubular shape with diameter bigger than 100nm. Figure 1 (d) shows the quantitative EDX analysis of the sample compositions coated with gold, yielded C 50.51%, O 8.27% and Zn 22.59% which confirms the presence of ZnO on the CNTC. The presence of Fe contributes around 2% of the composition due to the iron catalyst used in synthesis of CNT from WCPO.

The CNTC XRD pattern shows an intense peak at $2\theta = 25.9^{o}$ corresponding to the (002) reflection. There were also a weak peak (311) at $2\theta = 31.204^{o}$ which corresponds to a small quantity of iron catalyst during chemical vapor deposition process. CNTCZnO peaks at 26° and 43.8° corresponds to (002) and (101) planes of CNTC. Other peaks match well with the hexagonal wurtziteZnO. All the sharp peaks indicate the sample was well crystallized.

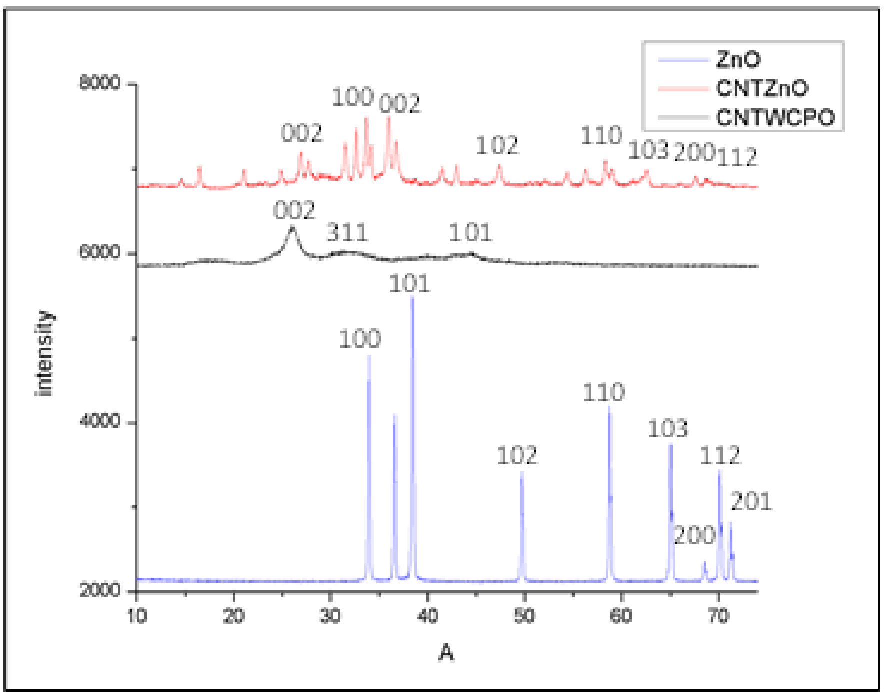

Figure 2: XRD diffraction patterns of ZnO, CNTC and CNTCZnO

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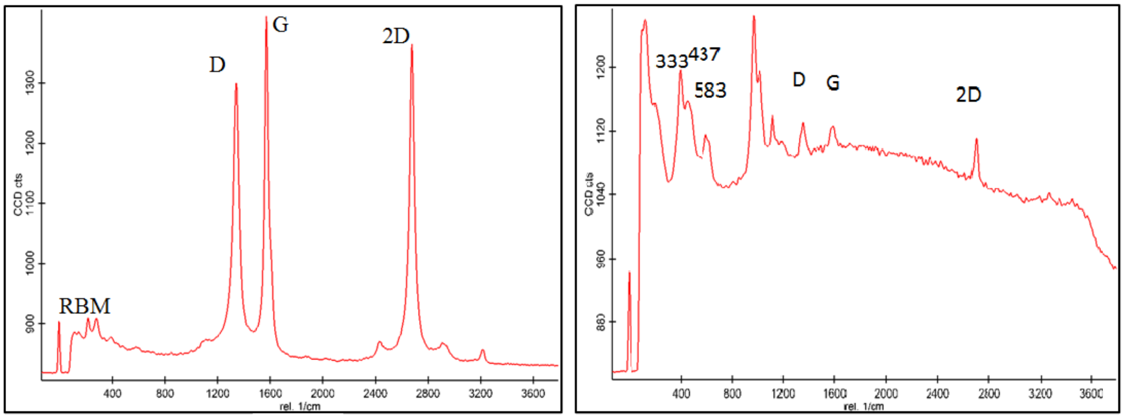

(a) *(b)*

Figure 4: Raman spectra of (a) CNTC and (b) CNTCZnO

The Raman spectrum of CNTC and CNTCZnO hybrid are shown in Figure 3. It can be observed from Figure 3(a) that the typical D, G and 2D bands of CNTs are present at 1331cm^{-1}, 1584 cm^{-1} and 2682 cm^{-1} respectively. It is believed that the CNTC is a mixture of single walled and multi walled carbon nanotubes due to the presence of additional radial breathing mode peak (RBM) at 213 cm^{-1} which confirms the presence of single walled carbon nanotubes (SWCNT) in CNTC. This gives a calculated diameter for SWCNTS at1.07nm which is relatively small as compared to the other multi-walled CNTs in the sample [6]. Meanwhile, in the CNTCZnO hybrid spectra as shown in Figure 3(b), the D and G band peaks of CNTC are clearly seen at their corresponding peaks. It is also observed that ZnO nanostructure peaks emergedat 333 and 437 cm^{-1} which is the characteristic of wurtzite hexagonal phase of ZnO.

CONCLUSION

CNT cotton has successfully synthesized from chemical vapor deposition process using waste cooking palm oil as the carbon source. Zinc oxide has successfully deposited on the CNT cotton (CNTC) using electrochemical deposition method. The diameter of CNTC which consisted of SWCNT and MWCNT increased after the coating of ZnO layer along its tubular shape. In this case, the CNTC presumably act as a nano electrical bridge between the zinc oxide nanoparticles for efficient charge mobility. Thus, the hybridization of CNT cotton which is mechanically strong, light weight, excellent conductors of heat, and transport electrons with zinc oxide is promising to be applied as flexible piezoelectric nanogenerator.

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