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The Effect of Graphitization Temperature on the Composition and the Electrical Conductivity of Carbon Nanotube

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Abstract

Carbon nanotubes (CNT) have been intensively investigated due to their superior electrical, thermal, and mechanical properties. In this work, CNT were synthesized using a relatively simple method that is catalytic graphitization. Catalytic graphitization was performed using bacterial cellulose as precursor, iron (III) chloride hexahydrate ($FeCl_3 \cdot 6H_2O$) as catalyst and chitosan as coupling agent and dispersant. Bacterial cellulose (cellulose source) was obtained by fermenting medium using acetobacter xylinum. Prior to usage, bacterial cellulose was chemically treated by soaking in a 0.5% chitosan solution for 2 hours at room temperature, followed by soaking in a 0,1 M catalyst solution at a temperature of 60 °C for 24 hours. The previous work did not variate the graphitization temperature, so in this work graphitization was conducted in a furnace under the flow of inert nitrogen gas atmosphere at 600 °C, 750 °C, 900 °C, and 1000 °C for 2 hours. CNT samples were characterized using Electron Dispersive Spectroscopy (EDS), Transmission Electron Microscopy (TEM) and LCR meter. The results indicate that the optimum catalytic graphitization temperature of CNT is 1000 °C which creates a bamboo-like CNT structure. It was also found that the electrical conductivity depends linearly on graphitization temperature. The highest electrical conductivity of 7.41×10^4 S/m is obtained for CNT sample synthesized at 1000 °C.

Keywords: Carbon nanotube, Catalytic graphitization, Bacterial cellulose, Chitosan, Electric conductivity

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1. Introduction

Since invented in 1991 by Sumio Iijima, carbon nanotube (CNT) has attracted significant interest of researchers due to their superior properties. CNT has a good chemical resistancy, good electrical and thermal conductivity, high surface area, and high mechanical

strength [1]. These properties open the possibility of using CNT for various application such as: transistor, battery, hydrogen storage, and supercapacitor [2].

Some conventional CNT synthesis methods like laser ablation, electric arc discharge, and chemical vapour deposition have been developed [3]. These methods produce high purity CNTs, but require rather high processing temperature and low pressure (vacuum) condition. Arc discharge requires temperature in the range of 5000-20000 ˚C while laser ablation uses temperature of 4000-5000 ˚C [2]. High temperature means higher production cost that will inhibit their use for mass CNT production. Therefore, less complex process and low cost alternative method is urgently needed [2].

The production cost can be reduced by decreasing the processing temperature. The processing temperature can be decreased from 2000-2500 ˚C to below 1000 ˚C by using transition metals or inorganic materials as a catalyst. This method is well known as catalytic graphitization [4]. To synthesize CNT with this method one needs carbon precursors, catalyst from metals and a coupling agent. In this research, bacterial cellulose was used as carbon precursor, Fe metal derived from $FeCl_3$ as catalyst, and chitosan as coupling agent. The bacterial cellulose was chosen instead of plant cellulose because it has higher purity, crystallinity degree, tensile strength and elastisity [5].

Bacterial cellulose used in this work was obtained from raw nata de coco. The cellulose is an outcome of bacterial fermentation in the medium containing carbon as the main constituent element. The $FeCl_3$ catalyst was used because it is low cost, easily obtainable, and more stable than any other iron complexes [4-6].

This research is aimed at optimizing a previous work [3]. Here, we focused on the effect of graphitization temperature on the composition and the electrical conductivity of CNT. In the previous work, the processing temperature was kept constant and the electrical conductivity of the synthesized CNT was not measured. The graphitization temperature is expected to affect the structure and electrical conductivity of CNT.

2. Material and Methods

2.1. Materials and instruments

Materials used in this work are bacterial cellulose from nata de coco, $FeCl_3.6H_2O$, chitosan, and NaOH. Bacterial cellulose, used as a carbon source, was obtained from Nata Lima Bersaudara factory in Padang, $FeCl_3.6H_2O$ (Merck), chitosan was made into powder from the waste of crustacean shell, and NaOH solution. Instruments used in this work are an electric furnace, an SEM SU 3500, a TEM JEOL JEM 1400, and a multimeter.

2.2. Methods

This work based on previously reported work [3]. Bacterial cellulose was washed using running tap water until reaching a neutral pH using pH meter to reduce its acidity and to remove its sugar content. Clean bacterial cellulose was boiled in a 1 M NaOH solution for an hour to eliminate impurities in cellulose. To remove its alkalinity, the cellulose was neutralized with runing tap water until reaching pH 7. The cellulose was cut into a box shape with a dimension of 2.5 cm x 2.5 cm x 1 mm. 0.5% w/v chitosan solution used as a coupling agent and dispersant was made by dissolving chitosan powder (0.5 g) in a 100 mL solvent. The solvent consists of 99.5 mL aqua DM and 0.5 mL acetic acid. A solution of $FeCl_3.6H_2O$ 0.1 M was used as a catalyst. To improve its homogeneity, catalyst solution was ultrasonicated using Branson 3510 Ultrasonic bath in 30 minutes. The cellulose was immersed in chitosan solution for 2 hours at room temperature and then added into the catalyst. 30 minutes sonication and 24 hours aging at 60 °C in an oven were carried out to enhance the absorption of the catalyst. Samples were dried in the oven at 40 °C for 3 hours. Graphitization process was performed in a furnace at various temperatures that were 600, 750, 900, and 1000 °C for 2 hours. Nitrogen gas with a flow rate of 0.1 Nl/minute was flowed into the furnace.

The elemental composition of the resulting CNT was determined using an energy dispersive X-ray spectrometry (EDS). The microstructure and size of the resulting CNT were investigated using transmission electron microscopy (TEM). LCR meter was used to measure electrical resistance of the samples and then the electrical conductivity was calculated from the measured resistance with 2 mm thickness and 15 mm area of the sample, by using equation (1).

$$\sigma = \frac{L}{R\ A} \tag{1}$$

3. Result and Discussion

Fig. 1 shows the CNT synthesized using catalytic graphitization of bacterial cellulose. Each sample was labeled as A, B, C and D for 600, 750, 900 and 1000 °C graphitization temperature respectively.

Fig. 1 shows that there is no significant difference between sample A, B, and C, but sample D looks more burnt than the other samples. The carbon content of the CNTs was determined using EDS. The EDS result is listed in Table 1. The carbon (C) content increases from sample A, B and D but it decreases in Sample C before reincreases at sample D. It is likely because sample C has the highest iron (Fe) content. When the

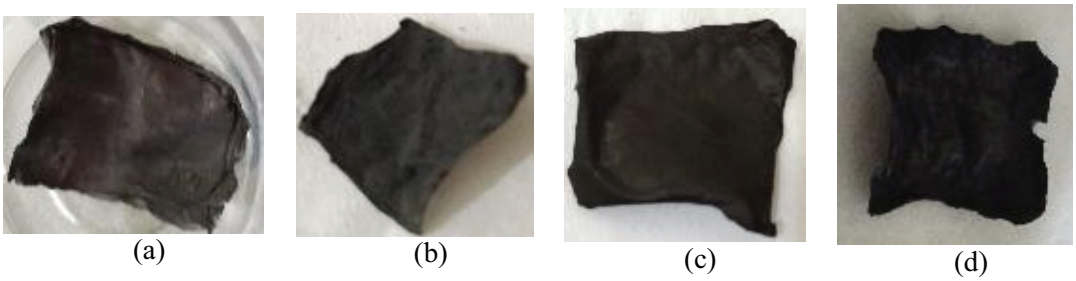

Figure 1: Bacterial cellulose sheet after graphitization.

temperature increases so the iron content decrease, it means the iron have done its role as catalyst well. Sample D with 1000 °C graphitization temperature has the highest carbon content and the lowest iron content. The sample D was used for TEM analysis.

TABLE 1: Carbon and other elements content of the synthesized CNT.

Elements	% Atomic			
	Sample A	Sample B	Sample C	Sample D
C K	58.02	62.14	27.71	80.80
O K	38.31	19.37	10.38	13.01
K K	0.24	0.81	7.33	0.30
Fe K	3.44	3.80	43.09	0.65
F K	-	12.99	11.08	5.09
Si K	-	0.39	-	-
Cl K	-	0.31	-	-
Co K	-	0.18	0.01	-
P K	-	-	0.28	-
TaM	-	-	-	0.06
Ca K	-	-	0.16	-

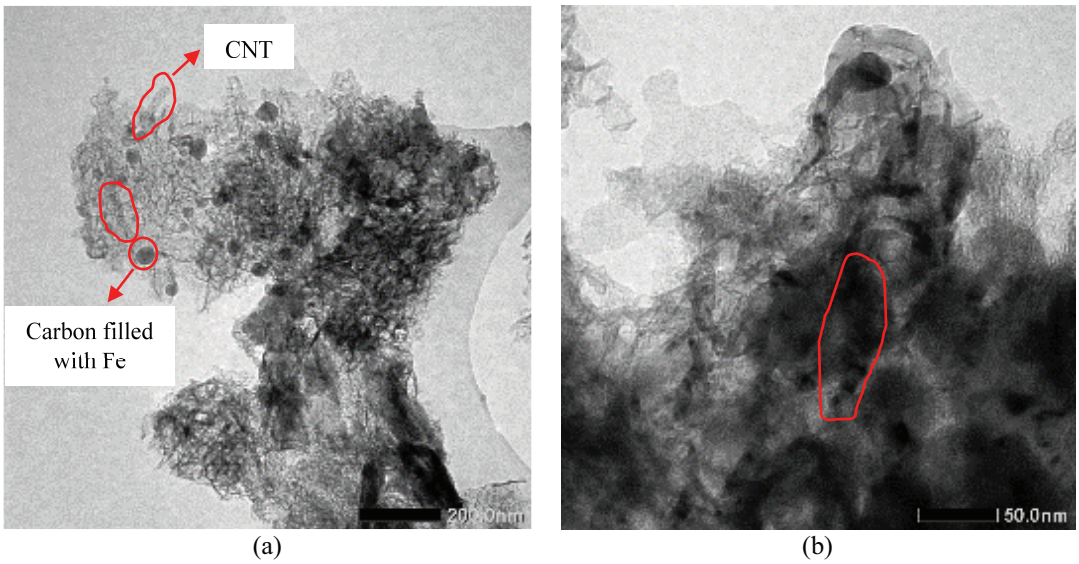

Figure 2: TEM images CNT sample synthesized using catalytic graphitization at 1000 °C.

Fig. 2 (a) shows that CNT was synthesized successfully. However it still contains carbon filled metal (Fe) impurity in some area. From Fig. 2 (b), the 1000 ˚C temperature formed Multi Walled Nanotubes (MWNT) with bamboo-like stuctures. The average length of the formed CNT is 95 $\pm$ 10 nm and the average diameter is 9.88 $\pm$ 2.48 nm. The bamboo-like CNTs have parabolic shaped layers stacked regularly. The bamboo-like CNTs contain dangling carbon atoms along the edges. The dangling bond affect their chemical properties. The bamboo-like CNTs may be useful for some application such as low-cost hydrogen storage, electrochemical capacitors and lithium ion battery [7].

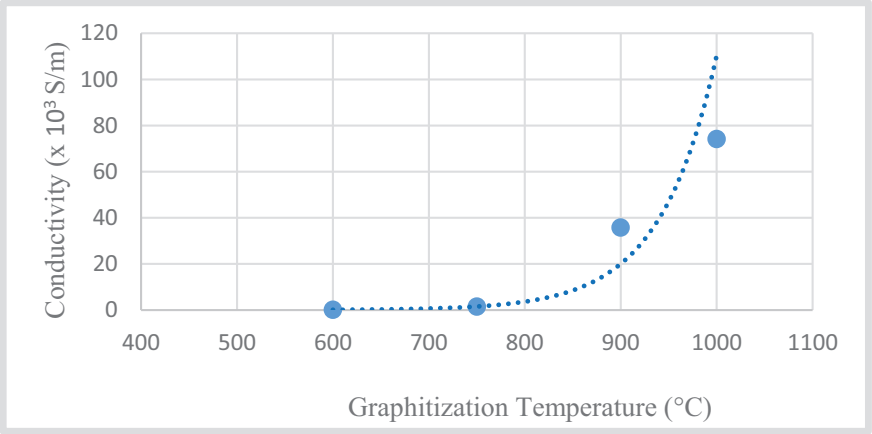

Figure 3: Electrical conductivity CNT samples.

Fig. 3 shows the result of conductivity in four samples in this work. Electrical conductivity increases almost exponentially with the graphitization temperature. The graph shows that conductivity increases with graphitization temperature. The highest conductivity of the synthesized CNT is 7.41x10^4 S/m at 1000 ˚C (sample D).

4. Conclusions

We can conclude that the optimum temperature to synthesize CNT with catalytic graphitization method is 1000 ˚C. The optimum temperature produces MWNT with bamboo-like structure. The electrical conductivity increases with graphitization temperature. The highest electrical conductivity 7.41x10^4 S/m is obtained for CNT sample synthesized at 1000 ˚C.

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