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Vertically aligned carbon nanotubes synthesized from waste chicken fat

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ABSTRACT

For the first time, vertically aligned carbon nanotubes (VACNTs) have been successfully synthesized using waste chicken fat as the starting material. Chicken fat oil, which was obtained through a rendering process, was directly mixed with 5.33 wt% ferrocene as a catalyst to form the synthesis stock. A mixture of single- and multi-walled VACNTs was synthesized at a fixed temperature of 750 °C in a thermal chemical vapour deposition furnace. Field emission scanning electron microscopy, micro-Raman spectroscopy and thermogravimetric analyses showed that the produced VACNTs were of excellent quality, comparable to those obtained using conventional carbon sources, with a purity of 88.2% and tube diameters ranging from 18 to 78 nm. Based on our study, waste chicken fat is a promising carbon source for the synthesis of high-quality and high-purity VACNTs.

1. Introduction

The production of carbon nanotubes (CNTs) using alternative sources has received much attention over the past 10 years. This increased focus has arisen because fossil fuel precursors are becoming more expensive, as the supply is predicted to run low over the next few decades. Innovative approaches using vegetable oils such as turpentine [1,2], eucalyptus [3], coconut [4], neem [5], palm oil [6–8] and waste cooking palm oil [9] as starting materials in CNTs production have been widely reported. The high carbon content in vegetable oils can produce large amounts of high-quality CNTs with purities reaching 90%. Palm oil, which has a chemical structure of $C_{55}H_{100}O_6$ [9], has a low C:H ratio of 1:2, compared to that of methane gas. The low H content in the precursor is expected to produce high-quality CNTs due to the low probability of by-product production, including amorphous carbon (a-C). This discovery has resulted in efforts extending to animal oils such as waste chicken fat. The selection of waste chicken fat as an alternative source for VACNTs growth is due to several important factors: (1) chicken fat has a high C content and a low C:H ratio due to the similarity between its chemical structure and that of vegetable oil, as it comes from the same lipid group; (2) waste chicken fat is a free source of carbon because it is usually discarded during poultry processing; (3) the utilisation of chicken fat as a precursor for VACNTs production can help in managing the disposal of chicken fat waste; (4) chicken fat that is intentionally disposed of into rivers can contaminate water supplies and harm ecosystems; and (5) the usage of waste chicken fat can be diversified beyond its use as biodiesel and biogas to use as a precursor for VACNTs production. In addition, biodiesel from chicken fat has a high sulphur content, is easily solidified at lower temperature and has a lower stability relative to vegetable oil as a biodiesel. It is also not economical to produce electricity from waste chicken fat biogas. These factors indicate that waste chicken fat is a potential source for VACNTs production, similar to palm oil and other vegetable oils.

Hence, we propose utilising waste chicken fat as an alternative and green carbon source for the production of vertically aligned carbon nanotubes (VACNTs). To the best of our knowledge, this is the first attempt to use animal oils in the synthesis of VACNTs. The synthesis was carried out using the catalytic thermal chemical vapour deposition (TCVD) method. Chicken fat and skin were collected from the wet market and then converted into a carbon precursor through a rendering process.

2. Materials and methods

Chicken fat, including the skin, was collected from a wet market. To prepare the precursor, the chicken fat underwent a

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rendering process. In this process, the fat was heated to 200 °C to convert the fat into an oily liquid. The oil was then filtered to separate it from the solid fat and the charred remnants of the chicken skin. Ferrocene was used as a catalyst and mixed directly with the oil at 5.33 wt%. The oil–catalyst mixture was stirred thoroughly for 30 min, and 6 ml of the mixture was then introduced into the precursor furnace of a TCVD system. The substrate was a mirror-polished p-type (100) silicon (Si) wafer 1×1 cm^2 in size which was ultrasonically cleaned in acetone. The oxide layer was etched from the wafer using a diluted solution of HF before rinsing in DI water. Next, it was air dried and then placed at the centre of the synthesis furnace which was set at 750 °C (deposition temperature). The precursor furnace was set to 470 °C according to the vapourisation temperature of chicken fat, which was initially determined using thermogravimetric analysis (TGA; Fig. 1(a)). The experiment was carried out for 60 min under an Ar atmosphere. The product deposited on Si substrates was collected and analysed by field emission scanning electron microscopy (FESEM-Hitachi SU8020), high-resolution transmission electron microscopy (HRTEM-JEOL JEM 2100), micro-Raman spectroscopy (Renishaw InVia microRaman System) and TGA (Perkin-Elmer Pyris 1).

3. Results and discussion

Fig. 1(b)–(d) shows FESEM images of the VACNTs obtained from the thermal catalytic decomposition of waste chicken fat. VACNTs 35 μm in length were measured, as shown in Fig. 1(b), representing an approximate growth rate of 0.58 μm min^{-1}. The FESEM images also show that the VACNTs arrays were fairly clean, except for the presence of a few small CNTs observed at several places, which attached to the surfaces of some VACNTs. The produced VACNTs had diameters ranging from 18 to 78 nm. This observation was supported by HRTEM images (Fig. 1(e)), wherein the observed outer diameter was 18 nm, including an inner layer of 4 nm and consisting of approximately 18 layers.

Micro-Raman spectroscopy was used to determine the crystallinity of the samples. The Raman analysis displayed in Fig. 2(a) shows two typical peaks at 1577.3 cm^{-1} for the G peak and 1346.3 cm^{-1} for the D peak. The ratio of the intensity of these peaks, I_D/I_G, was found to be 0.63, indicating a reasonable crystalline quality. The radial breathing mode (RBM) peaks, indicating the presence of single-walled CNTs (SWCNTs) in the sample, appeared in the range of 200–400 cm^{-1}, as shown in Fig. 2(b). The tube diameters, d, of the SWCNTs can be

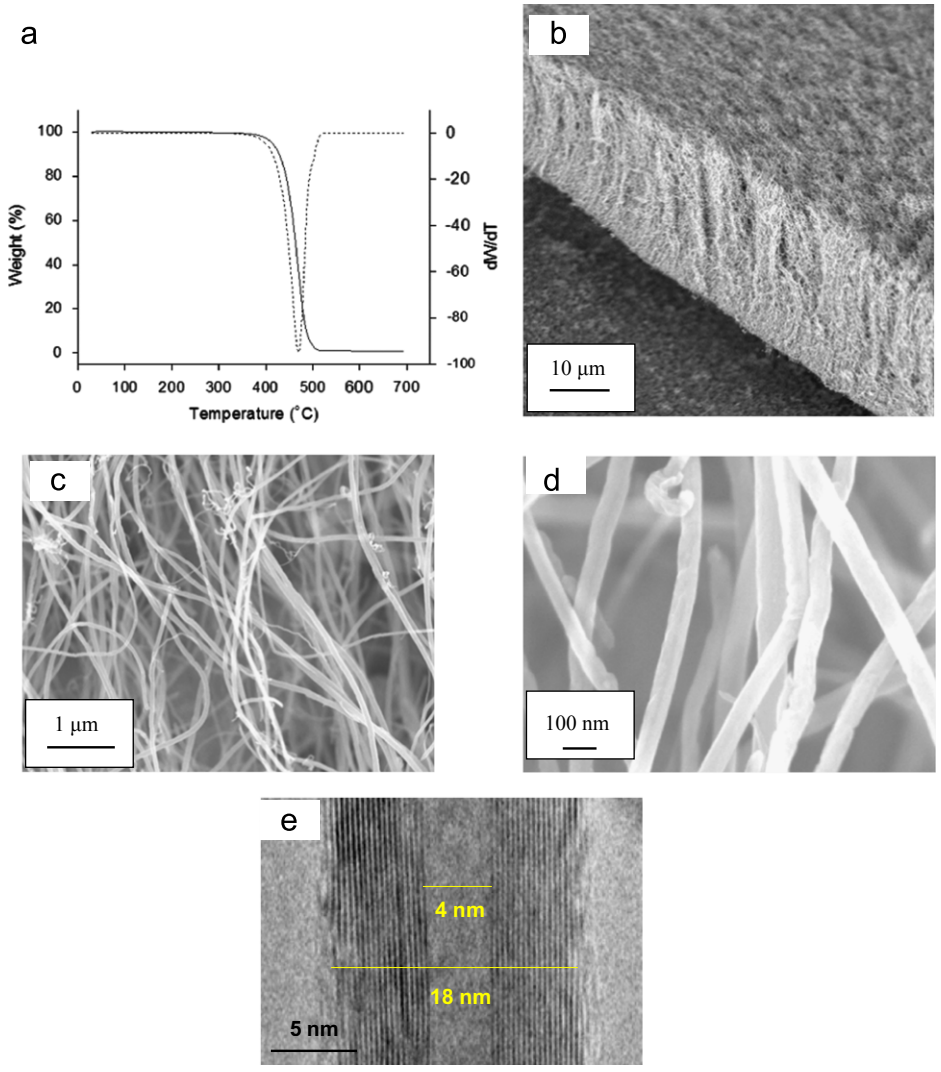

Fig. 1. (a) TGA and DTGA curves for chicken fat oil. (b)–(d) FESEM images of VACNTs grown from waste chicken fat on a Si substrate with increasing magnification. (e) HRTEM image of multi-walled CNTs.

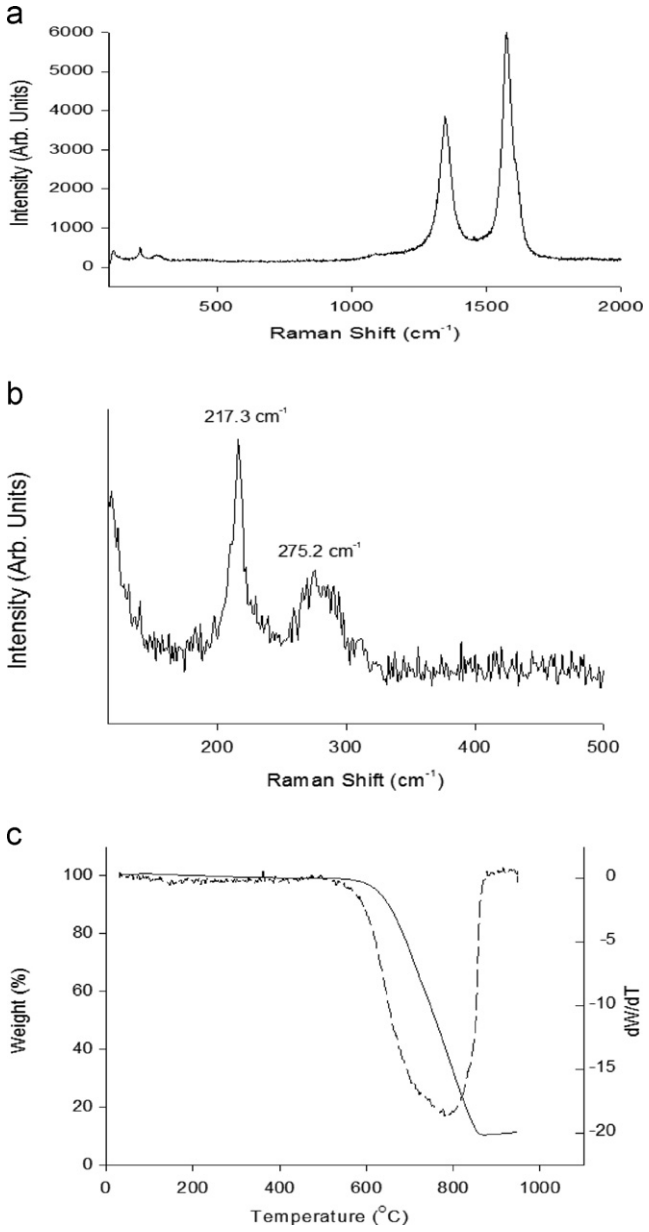

Fig. 2. (a) Micro-Raman spectrum showing the RBM modes and the D and G lines of the VACNTs, (b) a close-up image of the micro-Raman spectrum showing the RBM modes of SWCNTs, and (c) TGA and DTGA curves of VACNTs grown with waste chicken fat.

calculated from the RBM peak frequency, ω, where $d = 248\ (\mathrm{cm}^{-1})/\omega\ (\mathrm{cm}^{-1})$ [10]. In this sample, the RBM peaks appeared at 215 and 279 cm^{-1}, corresponding to SWCNTs diameters of 1.2 and 0.9 nm for the outer and inner diameters, respectively.

Fig. 2(c) shows a TGA and DTGA graph for the as-grown VACNTs. An initial weight loss of 0.3% was detected at a temperature of 110–250 °C. This loss was due to the decomposition of hydrocarbon impurities. The burning that occurred from 250 to 550 °C was mainly due to the presence of a-C in the VACNTs [11]. The a-C constituted approximately 1.3% of the total weight, indicating a minimal a-C content in the produced VACNTs. The significant weight loss indicates burning of the VACNTs, which ocurred from 642 to 880 °C. It should be noted that 10.6% of the sample weight remained after performing TGA up to 880 °C. This result was mainly due to the presence of Fe and other non-volatile constituents found in the VACNTs. From the TGA analysis, the purity of the produced VACNTs was found to be approximately 88.2%, and the sample can therefore be considered to be of high-quality.

The conventional CNTs growth mechanism was used to investigate the synthesis process of the VACNTs produced from waste chicken fat. Heating the precursor furnace to 470 °C resulted in in-situ Fe catalyst deposition in the synthesis region, which was set to 750 °C. At this temperature, chicken oil molecules instantaneously decomposed into lighter hydrocarbon precursors before catalytically decomposing on the Fe surface, leaving only the carbon atoms required for VACNTs growth along with the release of hydrogen and oxygen atoms. The hydrogen and oxygen atoms may form OH radicals that can act as a strong etchant to suppress the supersaturation of C–H molecules at the growing edge:

$$OH^{\bullet} + CH \rightarrow C + H_2O \quad (1)$$

Accordingly, the quality of the VACNTs was improved with minimal a-C content with the presence of oxygen in the waste chicken fat precursor. The presence of water vapours was observed within the tube furnace, which is consistent with the proposed equation. Due to the Fe catalyst that initially settled onto the Si substrate, it is believed that the initial growth of the VACNTs was established via a bottom growth mechanism.

4. Conclusion

Good crystallinity (I_D/I_G ratio of 0.63) and high-purity (88.2%) VACNTs with minimal a-C content were successfully synthesized from waste chicken fat. The obtained VACNTs were a mixture of single- and multi-walled CNTs. Waste chicken fat is a free and green carbon source alternative to the conventionally available fossil fuel precursor. The production of VACNTs from waste chicken fat involves a very simple method that can be easily scaled up for mass production. This project offers a solution to the chicken fat environmental pollution of ecosystems and represents a new application of chicken fat in the field of nanotechnology. If the control parameters are carefully chosen, it may be possible to produce VACNTs with excellent crystallinity and high purity at industrial volumes. Hence, waste chicken fat is a promising new carbon precursor source for next-generation VACNTs production.

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References

[1] Afre RA, Soga T, Jimbo T, Kumar M, Ando Y, Sharon M, et al. Carbon nanotubes by spray pyrolysis of turpentine oil at different temperatures and their studies. Microporous Mesoporous Mater 2006;96:184–90.
[2] Ghosh P, Soga T, Afre RA, Jimbo T. Simplified synthesis of single-walled carbon nanotubes from a botanical hydrocarbon: turpentine oil. J Alloys Compd 2008;462(289):93.
[3] Ghosh P, Soga T, Afre RA, Jimbo T. A simple method of producing singlewalled carbon nanotubes from a natural precursor: eucalyptus oil. Mater Lett 2007;61:3768–70.
[4] Paul S, Samdarshi SK. A green precursor for carbon nanotube synthesis. New Carbon Mater 2011;26:85–8.
[5] Kumar R, Tiwari RS, Srivastava ON. Scalable synthesis of aligned carbon nanotubes bundles using green natural precursor: neem oil. Nanoscale Res Lett 2011;6:92.

[6] Suriani AB, Azira AA, Nik SF, Md Nor R, Rusop M. Synthesis of vertically aligned carbon nanotubes using natural palm oil as carbon precursor. Mater Lett 2009;63:2704–6.
[7] Suriani AB, Salina M, Sarah MSP, Md Nor R, Anuar A, Siran YM, et al. The effect of precursor vaporization temperature on the growth of vertically aligned carbon nanotubes using palm oil. Defect Diffus Forum 2011;312:906–11.
[8] Azmina MS, Suriani AB, Falina AN, Salina M, Rosly J, Rusop M. Preparation of palm oil based carbon nanotubes at various ferrocene concentrations. Adv Mater Res 2012;364:408–11.
[9] Suriani AB, Md Nor R, Rusop M. Vertically aligned carbon nanotubes synthesized from waste cooking palm oil. J Ceram Soc Jpn 2010;118:963–8.
[10] Saito R, Dresselhaus G, Dresselhaus MS. Trigonal warping effect of carbon nanotubes. Phys Rev B 2000;61:2981–90.
[11] Chen CM, Chen M, Leu FC, Hsu SY, Wang SC, Shi SC, et al. Purification of multiwalled carbon nanotubes by microwave digestion method. Diamond Relat Mater 2004;13:1182–6.