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Synthesis, structural, and field electron emission properties of quasi-aligned carbon nanotubes from gutter oil

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HIGHLIGHTS

- Gutter oil was used as starting material to synthesise CNTs by TCVD method.
- CNTs of good quality (I_D/I_G ~ 0.66 and purity ~ 81%) were successfully produced.
- The synthesised CNTs show a potential for field electron emission application.

GRAPHICAL ABSTRACT

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ABSTRACT

Quasi-aligned carbon nanotubes (CNTs) have been successfully synthesised from the simple pyrolysis of gutter oil as starting material and ferrocene as a catalyst. The synthesis process was performed at synthesis and vaporisation temperatures of 800 and 250 °C, respectively, in a thermal chemical vapour deposition furnace. The CNTs synthesised using gutter oil have an overall diameter of about 30–50 nm, length of 30 μm, I_D/I_G ratio of 0.66, and purity of 81%, comparable to those obtained using conventional carbon sources. A field electron emission study of the CNTs exhibited a low turn-on and threshold field of 1.94 and 2.94 V μm^{-1}, which corresponded to current densities of 100 $\mu A\ cm^{-2}$ and 1.0 $mA\ cm^{-2}$, respectively which indicate that the CNTs synthesised are suitable candidates for use as field electron emitters. The synthesised CNTs from gutter oil also open up potential mass production applications in energy storage devices. This study demonstrates that gutter oil, a low-cost and readily available resource, can be used as an inexpensive carbon source for the mass production of CNTs.

1. Introduction

In recent years, a great deal of research effort has been devoted to the production of carbon nanotubes (CNTs) using low cost and readily available carbon precursor from waste resources such as waste plastic [1], scrap tyre rubber [2], waste glycerol [3], and waste engine oil [4]. These efforts have arisen due to the high cost of petroleum-based precursors (such as methane [5], acetylene [6],

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and ethylene [7] and [8]) as the supplies are predicted to be depleting in next few decades. Prior to the use of waste-based material as an alternative source of carbon, crop-based carbon sources including camphor [9–11], turpentine [12], eucalyptus [13], palm [14–17], coconut, olive, and corn oil [18] have been utilised to synthesise CNTs. Although the use of these green and renewable carbon sources seems to be appealing, their use for carbon precursors would leads to food shortage, increase the price of food products, and may contribute to environmental damage as they are grown on land converted from rainforests, peatlands, savannas, and grasslands [19]. These problems indicate that the crop-based oils may not be the only future resources for the production of CNTs.

Previous studies have shown that food-based waste such as waste cooking palm oil [20] and waste chicken fat [21–23] have been utilised for the synthesis of CNTs. These syntheses are possible due to the high carbon content originating from the fatty acids contained in the structures of both the oil and the fat. The utilisation of food-based waste precursor can now extend to gutter oil extracted from fat, oil, and grease (FOG) deposited in sewerage system. Gutter oil is a promising carbon source as it consists of a mixture of carbon-rich hydrocarbons. The gas chromatography mass spectrometry (GC-MS) and CHNS analyses in Table 1 and Table 2, respectively confirm that gutter oil has high carbon content and low C:H ratio, ideal to produce good-quality CNTs due to the low probability of by-product formation such as amorphous carbon [21]. In addition, the presence of oxygen in the structure will oxidise the amorphous carbon in-situ [24]. The high level of carbon content in the gutter oil is the main contributing factor that makes gutter oil the suitable precursor for mass production of CNTs.

The utilisation of gutter oil as carbon source not only can reduce the production cost of CNTs, but also help to manage the disposal problem of FOG. Conventional treatment of FOG such as land filling, land application, and incineration are not economical and associated with soil and air pollution problem. Alternatively, FOG are recycled for various uses including biodiesel [25], biogas [26] and also sold as cooking oil at below-market rates [27]. Although recycling is considered to be eco-friendly, there is a significant technical challenge in converting the FOG to biodiesel where the traditional base catalysis does not work well with grease due to the formation of soap between the base and free fatty acid in FOG [25]. Meanwhile, converting the gutter oil into cooking oil can be hazardous and even fatal to human health as it contains large quantities of bacteria and other dirty elements that congregate in sewage and can cause diarrhoea as well as other serious health effects. Gutter oil is also highly toxic as it contains harmful substances such as fatty and trans fatty acids as well as strong carcinogens such as aflatoxins, which accumulate as a result of several uses before disposal. The fatty acids in gutter oil however can be useful in the production of carbon material, including CNTs.

Table 1
Compounds identified and yield (% area) by GC-MS of gutter oil.

Compounds	Retention time	Area (%)
Aromatic hydrocarbons		
Ethylbenzene	3.380	1.027
Total		1.027
Aliphatic hydrocarbons		
2-methyloctacosane	20.073	1.061
2-methyloctacosane	20.155	2.253
Tetratetracontane	20.266	3.023
Octadecanamide	21.053	0.861
Tetratetracontane	23.267	1.185
Total		8.383
Oxygenated hydrocarbons		
2-Pentanone, 4-hydroxy-4-methyl-	3.077	2.655
Tetrahydrofuran, 2,2-dimethyl-	3.864	4.031
2-Pentanone, 3-methyl-	4.114	2.258
Hydroperoxide, 1-ethylbutyl	4.376	0.777
Hydroperoxide, 1-methylpentyl	4.505	0.722
Cyclopentane, 1-acetyl-1,2-epoxy-	4.740	2.727
Octanoic acid	7.534	1.511
Nonanoic acid	8.918	1.791
Dodecanoic acid	12.766	1.807
Pentadecanoic acid	15.050	2.299
n-Hexadecanoic acid	17.171	62.580
Oleic acid	18.849	1.708
Octadecanoic acid	19.036	1.842
Total		86.708
Nytrogenated hydrocarbons		
Hexadecanamide	19.269	1.173
9-Octadecenamide, (Z)-	20.871	2.710
Total		3.883

Table 2
CHNS elemental analysis of gutter oil.

Element	C	H	N	S
Weight%	69.774	10.277	3.976	0.043

These factors indicate that the waste from improper disposal of FOG is a potential source for CNT production. To the best of our knowledge, this is the first attempt to use gutter oil in the synthesis of CNTs by thermal chemical vapour deposition (TCVD) method. The potential of the CNTs produced from gutter oil as a field electron emission (FEE) cathode and electrode material for supercapacitor devices were also studied.

2. Materials & method

Gutter oil is an extract of waste FOG collected from a grease trap of a food service establishment located in a sewerage system. FOG was heated to 150 °C to extract the oil with dark brown in colour. The extracted oil was filtered to remove debris. The EDS analysis of the gutter oil (Fig. 1 (a)) shows the presence of C, O and metallic components of Na and Ca. The high weight % of C in gutter oil is consistent with the CHNS analysis (Table 2). The detail analysis of the gutter oil using inductively coupled plasma-optical emission spectrometry (ICP-OES) was carried out to support the presence of metallic components performed by EDS. The measurement was repeated for three times to obtain the average value of the metallic components. In contrast to EDS, Fe and K were also detected together with Ca and Na. However, the concentrations of Ca, Na, Fe, and K were found to be relatively lower than the values reported by others [28] (Table 3). The low metal concentration detected in the FOG was due to the origin of the FOG. The FOG used in this study was collected from restaurant, which the amount of metal components were known to be low as they came from the food grease trap resource. The Na in the gutter oil originated from the cooking salt (NaCl) used in the cooking process [20] while Ca was presumably released from the concrete surfaces under low pH conditions [29]. In order to synthesis CNTs, 5.33 wt% ferrocene was added to the gutter oil extracted and mixed thoroughly for 30 min. The CNT synthesis process followed a technique reported previously [21]. Briefly, a two-stage tube furnace was used, where the gutter oil-ferrocene mixture was place in one stage and substrates of *p*-type (100) silicon in the other. The synthesis temperature was set at 800 °C and the vaporisation temperature was set at 250 °C according to the vaporisation temperature of the gutter oil, determined previously using thermogravimetric analysis (TGA) (Fig. 1 (b)). The CNTs samples produced were characterised using field emission scanning electron microscopy (FESEM-JEOL JSM 7001F), high resolution transmission electron microscopy (HR-TEM-JEOL JEM 2100F), micro-Raman spectroscopy (JASCO-NRS 1500W), and