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STUDIES ON THE SUITABILITY OF ALUMINA AS BIMETALLIC CATALYST SUPPORT FOR MWCNTs GROWTH IN A CVD REACTOR

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ABSTRACT

The role of alumina (Al_2O_3) as catalyst support for improving the selectivity and uniformity in carbon nanotube growth via Chemical Vapour Deposition (CVD) technique has been widely explored. In this study, the suitability of bimetallic (Fe-Ni)/ Al_2O_3 catalyst prepared via wet impregnation technique for the synthesis of multi-walled carbon nanotubes (MWCNTs) has been investigated. The surface morphology, thermal properties and the crystallinity of the bimetallic catalyst supported on alumina and Multi-Walled Carbon Nanotubes (MWCNTs) were characterized using various analytical techniques such as HRSEM, HRTEM, EDS, SAED, TGA-DTA, and BET surface area. The results revealed that the synthesized catalyst was crystalline with surface area of $286 \text{ m}^2/\text{g}$ as determined by BET equipment. The bimetallic (Fe-Ni) supported alumina catalyst grown in a horizontal CVD reactor was found to produce high quality MWCNTs with 79.5 % purity. A Dynamic Light Scattering (DLS) based correlation chart revealed interdependence of length, diameter and aspect ratio of the purified and un-purified MWCNTs. This study has demonstrated that a thermally stable and high quality MWCNTs can be obtained from Fe-Ni/ Al_2O_3 catalyst via wet impregnation followed by CVD technique.

Keywords: Iron-nickel catalyst; Alumina support; MWCNTs; Characterization; DLS-Correlation

1. INTRODUCTION

The synthesis and application of carbon nanotubes (CNTs) have attracted considerable interest among experts in the field of nanoscience and nanotechnology due to their excellent electrical, mechanical and thermal properties (Hoenlin *et al.*, 2003; Konget *et al.*, 2000; Liu *et al.*, 2011). These remarkable characteristics make CNTs a suitable materials for various applications in hydrogen storage, nanoelectric devices, reinforced materials, energy storage, chemical sensors and field emission to mention but a few. Different methods of synthesising CNTs such as laser ablation, spray pyrolysis, arc discharge and catalytic chemical vapour deposition (CVD) have been reported in literatures (Ahmad *et al.*, 2013; Lee *et al.*, 2002).

However, of all the methods, CVD is considered rather simple, easy to scale up and a cost-effective technique in terms of production of carbon nanotubes of high quality yield and purity, better structural growth, and mass production compared to the other methods (Terrado *et al.*, 2006). Ease of optimization of the process parameters, feasibility of exploring numerous carbon sources, and its ability to run multiple samples per experimental run, also make a suitable process technique.

Over the years, there have been considerable increases in the utilization of CVD techniques for CNT synthesis based on the aforementioned advantages. Additionally, several approaches of producing CNTs including growing of metallic catalyst on support material have been



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reported (Pelechet *et al.*, 2009; Quianet *al.*, 2003; Yeohet *al.*, 2009). The choice of metallic catalyst with or without support materials influences the yield and morphology of synthesized carbon nanotubes. Not only that the mechanism of CNT growth is complex and not fully understood (Kumar, 2012). It is also noteworthy to mention that, catalyst design, catalyst type, support material as well as their properties are integral components in the synthesis of controlled-growth of carbon nanotubes (CNTs) for various applications (Mhlanga *et al.*, 2009). The role of catalyst support in supported-catalyst lies in its ability to determine accessibility of active sites, and to influence certain properties such as the pore volume and pore-size distribution which are essential parameters in catalyst design (Storchet *al.*, 1998). Thus, the efficiency of the CVD method for CNT growth is a function of catalysts preparation and metals loading (Terredo *et al.*, 2006).

Furthermore, Iron, Cobalt and Nickel are mostly employed as an efficient catalyst for CNT growth in the CVD method. The choice of these metals as active component of catalyst is ascribed to their ability to form carbides which are metastable compounds that decompose to form a graphene-like sheet by the process called graphitization. Thus, the nature of metallic catalysts as well as the support material or template both affects the yield and purity of the final CNT material (Kathyayini *et al.*, 2008). In the synthesis of CNTs by CVD, the catalyst aids in the decomposition of the carbonaceous materials during thermal process in the quartz chamber of CVD. However, the purity of the CNT material is still a subject of debate, due to the fact that the as-synthesized CNT is usually accompanied by impurities such as fullerenes, amorphous carbons, crystallized graphite, support material and the metal (Kruusenberget *al.*, 2011). The level of impurities is known to depend on the choice of metal, support material and other growth parameters.

Different support materials such as Al_2O_3 , zeolite, MgO, SiO_2 , and CaCO_3 have been applied as a carrier in supported-catalysts for CNT (Kathyayini *et al.*, 2008; Willemset *al.*, 2000; Mao-Lin *et al.*, 2012 and Couteauet *al.*, 2003). Several researchers have reported high purity multi-walled carbon nanotubes obtained from Co-Mo supported MgO catalyst via CVD (Pelechet *al.*, 2009; Quianet *al.*, 2003; Yeohet *al.*, 2009). Awadallah *et al.* (2012) demonstrated the synthesis of CNTs prepared via CVD methods from Ni-Mo and Co-Mo supported on Al_2O_3 catalyst. Studies have shown that alumina supported catalysts for CNTs synthesis appeared more promising due to creation of a high surface area and mesoporous materials which promote catalysis (Kathyayini *et al.*, 2008). In addition, Al_2O_3 has good thermal stability and can be easily removed after the synthesis via acid purification.

In this present study, bi-metallic Fe-Ni catalyst supported on aluminium oxide were developed and utilised to prepare CNT through CVD method. The choice of Fe-Ni as an active part of the catalyst is due to their availability and cheapness (Uddin *et al.*, 2008).

2. METHODOLOGY

2.1 Material

All the chemicals used in this study are of analytical grade with percentage purity in the ranges of 98- 99.99 %. These chemicals include Nickel nitrate hexahydrate [$\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$], iron nitrate nonahydrate [$\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$], alumina [Al_2O_3], distilled water and concentrated hydrochloric acid [H_2SO_4] were supplied by Aldrich. The carbon source (acetylene) and carrier gas (argon) are also of analytical grade with 99.99% purity supplied by BOC Nigeria.



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2.1.1 Synthesis of Supported Bimetallic Catalyst

Wet impregnation method was employed for bimetallic catalyst preparation. Bimetallic catalyst (Fe-Ni/Al₂O₃) was produced using equal weight percentage of Fe and Ni supported on Aluminium oxide. This was prepared by dissolving 4.04 g of Fe (NO₃)₃.9H₂O and 2.91 g of Ni (NO₃)₂.6H₂O in 50 ml of distilled water. The solution obtained was then added to 8 g of Aluminium oxide and the mixture was allowed to age for 45 min under constant stirring. The resulting homogeneous slurry was dried at a temperature of 115 °C for 7 hrs. The product obtained was grinded and calcined in a static air furnace at 400 °C for 2 hrs. The calcined sample was allowed to cool and was later sieved through 150 µm sieve.

2.1.2. Synthesis of CNTs

In this study, a cylindrical tube reactor of length 1010 mm with internal and external diameter of 52 mm and 60 mm respectively and thickness of 4 mm was used. 1.0 g of the supported bimetallic catalyst (Fe-Ni/Al₂O₃) was weighed and spread evenly on a quartz boat placed at the central part of the horizontal tube. The heating rate, temperature, gas flow rates were maintained at the desired rate. The entrapped gases in the quartz tube were expelled using argon as the carrier gas at a flow rate of 30 ml/min. At a temperature of 750 °C, the flow of acetylene was released into the quartz tube of catalytic reactor at 100 ml/min for 45 min with immediate increment in the flow rate of the carrier gas (argon) to 200 ml/min. As the residence time (45 minutes) of the reaction was attained, the flow of acetylene was stopped and the argon gas was left flowing at 30 ml/min until the reactor cooled to room temperature. The sample was removed, weighed and analysed. The yield of the deposited carbon was therefore determined using Equation (1) (Yeoh *et al.*, 2009; Taleshi, 2012).

$$\text{Yield (\%)} = \left(\frac{W_2 - W_1}{W_1} \right) \times 100 \quad (1)$$



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Where W₁ is the initial weight of the catalyst before reaction and W₂ is the weight of catalyst and carbon deposited after synthesis.

2.1.3. Purification of As-synthesised Carbon Nanotubes

The as-produced CNT was treated with 30 % wt concentrated sulphuric acid, heated and stirred at a temperature of 60 °C for 30 minutes using a magnetic stirrer to remove residual fractions of Al₂O₃ or Fe-Ni in the form of sulphates. The sample was washed with distilled water until the pH was approximately 7.0 and later dried at 110 °C for 5 hours. The acid purified sample was characterized with SEM, TEM, EDS, SAED and TGA.

2.2 Characterization of the Synthesised Materials

2.2.1 Thermo-Gravimetric Analysis (TGA) and Differential Thermal Analysis (DTA)

The thermal stability, compositional and percentage purity of materials were determined using TGA 4000 (PerkinElmer). Samples were analysed in nitrogen environment at a flow rate of 20 ml/min, pressure of 2.5 bars and heating rate of 10 °C/min. To a zeroed thermal balance, sample was loaded and recorded into the equipment using pyris manager software. The analysis was then initiated after constant weight was noted using the created heating profile (temperature scan). The test results were then analysed using pyris manager for proximate and compositional analysis.

2.2.2 High Resolution Scanning Electron Microscope (HRSEM)

The surface morphology and microstructure of the synthesised materials were characterized using Zeiss Auriga HRSEM. The HRSEM equipped with EDS was further used to determine the elemental composition of the synthesised catalysts. A small quantity of the synthesised



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materials was sprinkled on a sample holder and sputter coated with Au-Pd using Quorum T150T for 5 minutes prior to analysis. The sputter coated samples were firmly attached to the carbon adhesive tape and analysed using Zeiss Auriga HRSEM equipped with In-lens standard detector at 30 kV. The microscope was operated with electron high tension (EHT) of 5 kV for imaging.

2.2.3 X-Ray Diffraction Patterns (XRD)

The crystal phase identification of the powdered materials were performed using Bruker AXS D8 X-ray diffractometer system coupled with Cu-K α radiation of 40 kV and a current of 40 mA. The λ for K α was 0.1541 nm, scanning rate was 1.5 °/min, while a step width of 0.05° was used over the 2 θ range value of 20 – 80 °.

2.2.4 High Resolution Transmission Electron Microscope (HRTEM)

The diameters and the crystalline nature of as-produced and purified CNTs were determined by Zeiss Auriga HRTEM operated at 3950 V. Small quantity of the materials were suspended in 10 ml methanol and ultrasonicated until the particles completely dissolved. Few drops of the slurry was placed onto the holey carbon grid, dried via exposure to photo light and analysed.

2.2.5 Particle Size Analysis (DLS Technique)

The particle size and the hydrodynamic diameter were determined using Zetasizer Nano S at scattering angle of 173 ° operating at 25 °C with equilibrating time of 120 secs. 1mg of the samples were dispersed in 10 ml of ethanol then transferred into a polystyrene cuvette using a syringe with 0.22 μ m filter coupled to it. This was then placed in the analysis stage of the equipment for analysis.

2.2.6 BET Surface Area

The surface area of the developed and the alumina were determined using a BET method in Nova e-series equipment. Samples were degassed at 250 °C for 4 hrs for moisture and removal. The degassed samples were then analysed for physisorption of the adsorbate (nitrogen) by

the adsorbent in liquid nitrogen environment on the surface.

3. RESULTS AND DISCUSSIONS

Figure 1 depicts the thermal stability of the alumina support and the bimetallic supported catalyst. The TGA examines the behaviours of materials as a function of percentage weight and derivative weight against temperature (Okpalugo *et al.*, 2006). At 800 °C, the percentage weight difference retained in the catalyst increased by 1.621 % compared to the alumina support. This is resulted from the presence of active catalyst (Fe and Ni) in the composition which raised the thermal behaviour of the alumina for thermal stability for CNTs growth. The results indicated improvement of the thermal stability of the prepared bimetallic catalyst compared to the raw alumina. The BET surface area of the developed bimetallic catalyst was determined to be 285.62 m²/g under nitrogen condition, with micropore volume of 0.1023 cc/g and micropore half pore width of 32.43 Å. This surface area is little less than those reported by Abassi *et al.*, 2014. This may be due to the presence of weak bond that exist between Fe-Al₂O₃ (Zhang *et al.*, 2006).

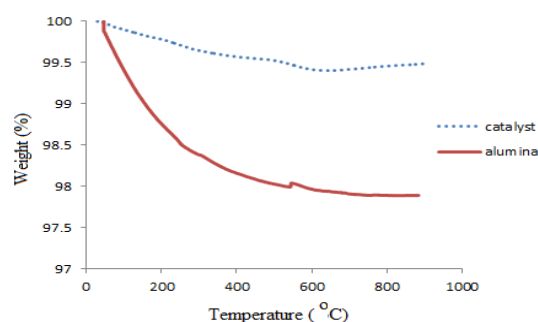


Figure 1: Thermal behaviour of the developed bi-catalyst (Fe-Ni/Al₂O₃) and raw alumina

The XRD pattern of the synthesized catalyst is presented in Figure 2 which demonstrates the presence of sharp diffraction peaks. This suggests that the prepared catalyst is highly crystalline, an indication of orderly distribution of the metallic ions on the pore of the alumina support. The characteristic peak at 2 θ value of 35.23° suggest the



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presence of alpha alumina ($\alpha\text{-Al}_2\text{O}_3$) (Kathyayini *et al.*, 2008). The crystalline size of the catalyst was calculated from the XRD data using the Scherrer Equation shown in Equation (2) (Chen *et al.*, 2006).

$$D = \frac{K\lambda}{\beta \cos\theta} \quad (2)$$

Where D is the particle size diameter, β is the full width at half maximum, λ is the wave length of X-ray (0.1541 nm), θ is the diffraction angle and K is the Scherrer constant (0.94).

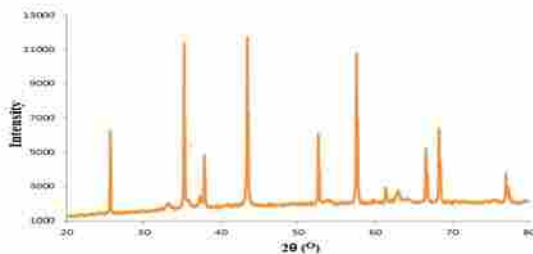


Fig.2: XRD spectra of Fe-Ni/Al₂O₃ catalyst

The particle size distribution of the synthesised supported catalyst based on the respective identified peaks in the XRD pattern is presented in Figure 3. The estimated crystallite size revealed that the bimetallic catalyst is dominated with particle sizes of 60.94 nm and relatively the smallest particle size population of 8.86 nm. Thus, the result indicates the possibility of producing CNTs of nano size when utilizing bimetallic supported catalyst.

The results presented in Table 1 showed that 45 minutes is the optimum reaction time for CNTs synthesis using bimetallic catalyst (Fe-Ni/Al₂O₃) at a desired temperature (750 °C), argon flow rate of 200 ml/min and acetylene flow rate of 100 ml/min. Below this reaction time, acetylene possesses low decomposition rate and little of the bi-metallic catalyst's active pores are susceptible to carbon nanotube yield (Hengameh, 2006). Further increment in the residence time from 45 - 60 minutes resulted into a slight decrease in CNTs yield from 49 % to 48 %. At 60 minutes reaction time, side-way reaction

occurs which resulted in catalyst deactivation and CNTs yield decrease with increase reaction time. The initial increment in the carbon yield may be attributed to continuous deposition of carbon from acetylene into the pore volume of the catalyst until the entire pores are completely saturated (45 minute). At 60 minutes, the catalyst becomes poisoning due to saturation of the catalyst pores by carbon deposit and further leads to reduction in yield.

Table I: Effect of Residence Time on Carbon Nanotube Yield

Time (mins)	Yield (%)	Production rate (g/mins)
15	25	0.0167
30	42	0.0140
45	49	0.0109
60	48	0.008

It has been reported that porous materials such as alumina are excellent catalyst support, but they have the tendency of producing amorphous carbon during CNTs production. However, wet impregnation of the bimetallic catalyst on the alumina will reduce the porosity of the support. This was confirmed by the SEM micrograph of the synthesized catalyst as presented in Figure 4. The HRSEM micrograph clearly showed that the synthesized catalyst is of low porosity with noticeable particles of metal ions covering the surface of the support material (Figure 4b). The influence of time on the quantity of carbon nanotubes yield was investigated and the results obtained are presented in Table 1.

Due to the possibility of presence of impurities as a result of undissolved catalyst particles, the synthesised carbon nanotubes were purified with 30% sulphuric acid to remove the undissolved metallic catalyst, support and amorphous carbon presence in the as-produced CNTs.

The carbon nano-material treated using this process becomes active and led to oxygenous functional groups generation such as carboxyl (Dalton *et al.*, 2000).

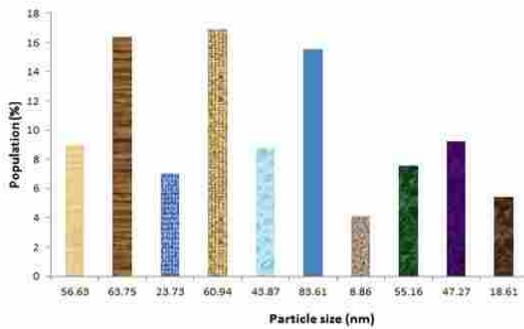


Fig.3: Percentage Population of Particle Sizes from XRD Data

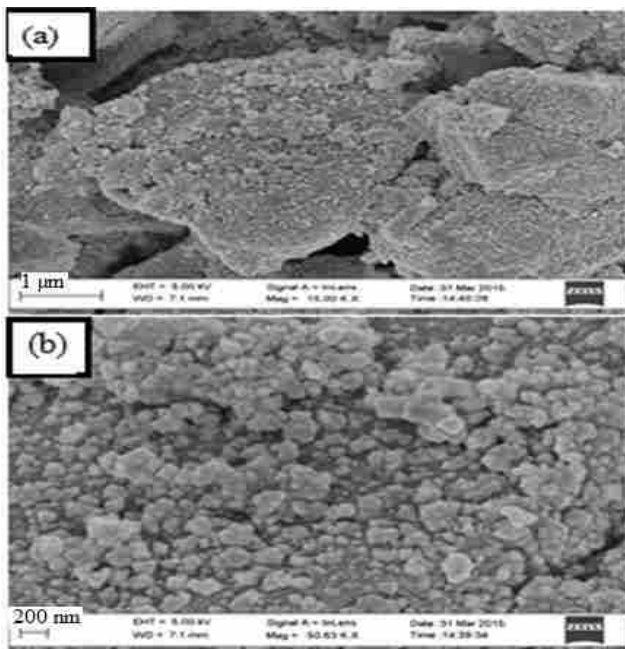


Fig.4: HRSEM Micrograph of (a) Low Magnification and (b) High Magnification of Calcined Fe-Ni/Al₂O₃ Catalyst

The HRSEM micrograph, Figure 5 shows a densely populated strand of CNTs with high degree of homogeneity covering the impurity present; which is in line with the property reported by (Ratkovic et al., 2011).

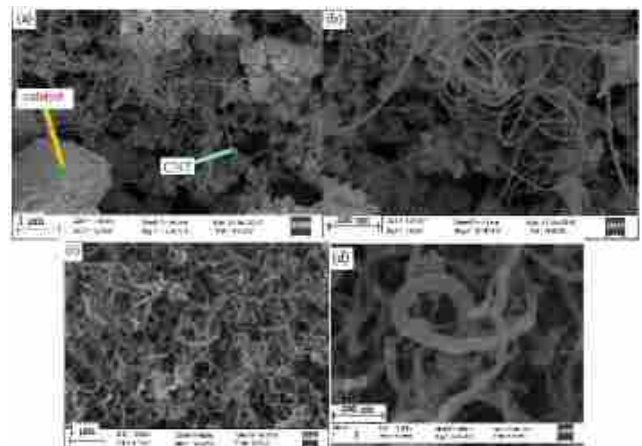


Fig.5: HRSEM Micrograph of As-Produced (a-b) and Purified (c-d) Carbon Nanotubes with Production Time of 45 Minutes

HRSEM micrograph of as-produced CNTs presented in Figures 5a and b indicate that the CNTs are several micrometres long with encapsulated catalyst impurities during CNTs nucleation process.

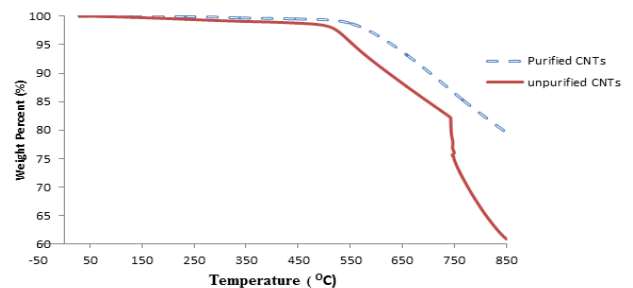


Fig.6: Thermal Behaviour of As-synthesized and Purified MWCNTs

Figures 6 and 7 represent the TGA and DTA profile of both the as-produced and purified CNTs respectively.

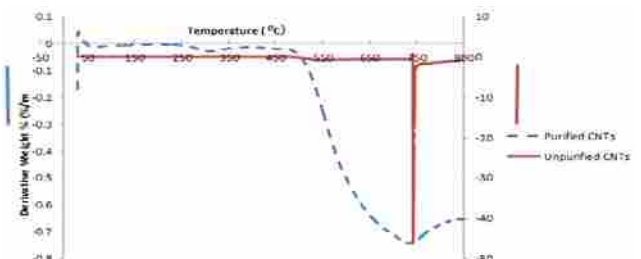


Fig.7: DTA of As-synthesized and Purified MWCNTs



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Additionally, Table 2 contains information about the sample extracted from Figures 6 and 7. The results indicate that the as-produced CNTs had lower moisture content than purified CNTs. The increment in percentage moisture content may be due to the purification process which involves distilled water and sulphuric acid solution, thus there is a possibility of water being entrapped within the pore of the CNTs. The TGA results also revealed that the percentage purity of the synthesized MWCNTs was 79.547 % with lesser volatile organic constituents compare to as-produced MWCNTs. The observable high T_p (optimum temperature of degradation) in unpurified CNTs is linked to the presence of impurities which accompany the synthesis CNTs (Ebbensen *et al.*, 1998; Eftehariat *et al.*, 2005). After the purification process, the impurities in the form of amorphous carbon and metals oxides were reduced and further lower the peak temperature of the purified carbon nanotubes. Temperature at which 50 % of the material degraded, T_{50} were also determined to be greater than 850 °C which is an indication of high thermal stability of the CNTs. Hence the CNTs produced with bimetallic catalyst, Fe-Ni on alumina support is thermally stable.

Table II: Proximate Analysis of the As-Synthesized and Purified MWCNTS 750 °C

Material	Moisture Content (%)	CNTs Content (%)	Volatile Content (%)	Degradation Temperature Range (°C)	T_{50} (°C)	T_p (°C)	Onset Temperature (°C)
As-produced MWCNTs	0.013	62.48	37.50	450.65-726.54	>850	743.64	516.71
Purified MWCNTs	0.098	79.55	19.87	519.80-806.91	>850	738.61	578.12

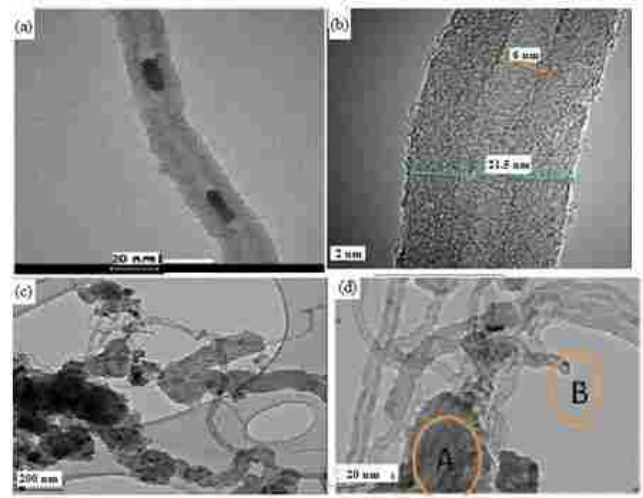


Fig.8: HRTEM Image of MWCNT (a) As-produced (b) Purified MWCNTs showing the Inner and Outer Diameter (c) As-synthesized and (d) Purified CNTs

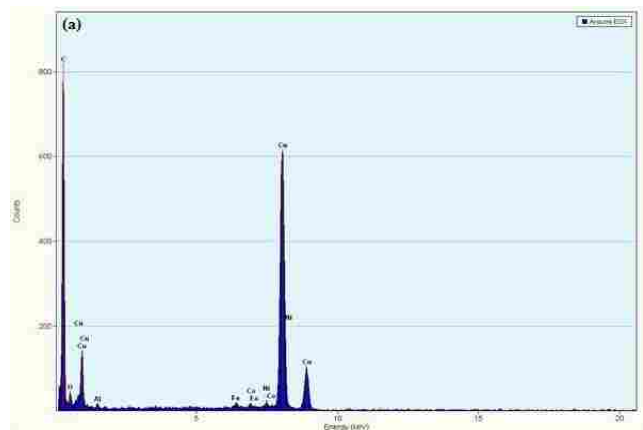


Fig.9: (a) EDS and (b) SAED of Purified Carbon Nanotubes

The morphologies and the microstructure of the unpurified and purified CNTs was examined using HRTEM and the results presented in Figure 8 a and b. According to figure 8a and b, the synthesised carbon nanotubes are multiwall



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carbon nanotubes (MWCNTs) having a straight length structure or rope-like structure with absence of coiled-tube. In the purified HRTEM image (Figure 8b), there was a reduction in the level of impurity present which was attributed to by the purification process by 30 % sulphuric acid. The acid aids in the precipitation of the metallic impurities in the form of amorphous carbon and metal carbide which might have not dissociate during the thermal catalysis process. The diameters and length of the microscopic images shown in Figure 7(a-d) are well represented in terms of their interrelationship in Figure 8. It can be deduced from the results presented in Figure 8c that there is no uniformity in the size distribution of the purified synthesized carbon nanotubes. However, point A shows some denser and thicker tubes which anchor some catalyst as a result of continuous overlapping and encapsulation therein with impurities of larger particles (Son *et al.*, 2008). The point marked B in Figure 8d indicates a cracked tube with thin tube end where catalysts particles were removed during the oxidative purification process. The EDS result presented in Figure 9 A also indicates that the metals impurities are not completely removed, hence, there is need to improve on the purification process. The presence of Cu in the EDS results emanated from the Cu grid used in the HRTEM analysis. The crystalline nature of the prepared MWCNTs was further evaluated via the Selected Area Electron Diffraction (SAED) pattern (Figure 9b). The SAED pattern revealed the presence of two broad crystal plane which correspond to (002) and (100) respectively. This result is in accordance with the previous studies (Conget *et al.*, 2011). The effect of acid treatment on the aspect ratio (L/D) of the purified carbon nanotubes was investigated by establishing a DLS-correlation chart. The diameter of as-produced and purified carbon nanotubes from HRTEM image was related to the D_h (hydrodynamic diameter) using modified Navier-Stokes equation (Nair *et al.*, 2008) and Stokes-

Einstein equation. The chart is aimed at determining MWCNTs aspect ratio and length at a specific diameter.

$$D_h = \frac{L}{\ln(L/d)+0.32} \quad (3)$$

Where D_h , l and d are hydrodynamic diameter, length and diameter of MWCNTs respectively. From the DLS-Nanosizer, the hydrodynamic diameter of both the as-synthesized and purified carbon nanotubes were determined to be 4847 nm and 3471 nm respectively.

From the correlation graph, Figure 10, the length and the diameter of as-synthesized MWCNTs purified with 30 % sulphuric acid shows a reduction in the diameter and the length.

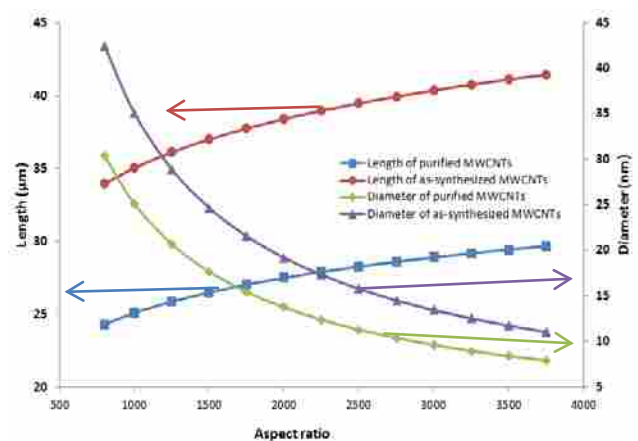


Fig.10: Aspect Ratio Correlation of Length and Diameter of Purified and As-Synthesized Long-Tube MWCNTs

This reduction in diameters and lengths confirms the removal of crystalline impurities in the as-synthesized MWCNTs. The aspect ratio (L/D) is proportional to the length and inversely proportional to the diameter of both the as-synthesized and purified MWCNTs. The correlation results also indicate a long-tube growth of purified and as-synthesized MWCNTs of 25.86 µm and 36.11 µm length respectively with 1250 aspect ratio. Based on the results of the analysis obtained, it can be concluded that bimetallic catalyst on alumina support is suitable for CNTs production.

4.0 CONCLUSION



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In this study, multi-walled carbon nanotubes were developed from bimetallic supported alumina catalyst via wet impregnation followed by CVD technique. High purity and thermally stable CNTs was successfully synthesised at optimum conditions of reaction time of 45 minutes, temperature of 750 °C, argon flow rate of 200 ml/min and acetylene flow rate 100 ml/min. The average diameter of the as-produced multiwalled carbon nanotubes obtained from bimetallic Fe-Ni/Al₂O₃ catalyst was 28.89 nm whereas; the bimetallic catalyst had average particle size of 46.24 nm. The correlation chart showed that the MWCNTs produced was a long-tube multiwall carbon nanotubes with aspect ratio of 1250. Acid treatment method with tetraoxosulphate (VI) acid of 30 % was found to remove reasonable amount of impurities with little tip-tube damage. Thus, the bimetallic catalyst on alumina support developed via incipient wet impregnation method is suitable for CNTs production by CVD method.

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