A NOVEL PREPARATION OF HIGH QUALITY SWCNTS FROM MARGOSA LEAVES FOR NANO PAPER BATTERY – A NEW FEASIBILITY STUDY

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ABSTRACT

A new modified AC method – VSA methodology (with KRS or NTFDS theory) was adopted in this present work for the preparation of CNTs from natural organics i.e., Margosa leaves towards possible application to nano electronics, nano paper batteries and nano computer system. Structural, Compositional, Surface Morphological and Nano structural Characterizations were carried out on harvested products. The effects of optimizations parameters like pH of the various dipping solutions (acidic, basic and neutral), volume of dipping solutions, various types and parts of the materials, various dipping timings, number of annealing and dipping, various annealing temperature, various time of annealing and various dipping solution temperatures on structural, compositional, surface morphological, nano-structural characterizations of materials and on high grade SWCNTs growth with high yield were studied intensively. Inferences from characterizations were derived and graphically emphasized. Correlation studies between these characterization inferences (such as grain size, purity) and above optimization parameters were carried out with a high light on yield of high grade SWCNTs. Beyond all of these, we have carried out a novel feasibility study at first time, which comprises the possible usage of precursor organic carbon sources i.e., margosa leaves for SWCNTs with high yield via a low cost technique and methodology as value in commercial efforts.

Keywords: *Modified AC method, VSA methodology, NTFDS theory, Natural organics, Margosa leaves, HRTEM, SWCNTs, Nano paper battery like energy storage system.*

1. INTRODUCTION

The present work showed that SWCNTs obtained by open air annealing and cooling of leaves of Margosa (i.e., neem tree). The present work made a break through via plays an alternative method to conventional, traditional methods such as arc discharge, laser ablation, metal catalysts, pyrolysis, and varieties of CVD methods and also removes the usage of synthetic chemicals [1-33]. The modified AC method [Zhenhui Kang et al, 33] – VSA methodology (with KRS or NTFDS theory) was adopted in this present work for the preparation of CNTs from natural organics i.e., margosa leaves. The entire steps such as Precursor materials selection with cleaning, Annealing, Sudden cooling, Interaction between red hot natural⇔organic carbon resource materials and Dipping solutions (DS) [including Nano Thermo Fluid Dynamics (NTFDS) and Nano-drilling process] involved in this process were explained.

2. EXPERIMENTAL DETAILS

A material i.e., leaves of Margosa (i.e., neem tree) collected from nature, cleaned in water and dried in open air. These materials were used without further purification. Then the materials were open air annealed (upto red hot) in a muffle furnace at various temperatures viz., 600^0C and 800^0C for various time of heating viz., 1 minute to 5 minutes. After that they were immediately dipped into various types of solutions viz., Sodium hydroxide (NaOH), Hydrogen peroxide (H_2O_2), Nitric Acid (HNO_3), Sulphuric Acid (H_2SO_4), Hydrochloric Acid (Hcl), Mineral Water (MW), Salt Water (SW), Double Distilled Water (D2W), Ice Water (Ice W), Hot Water (HW), Pure Water (PW), Ice water mixed double distilled water (IceW+D2W) and Hot water mixed double distilled water (HW+D2W) solution for various solution temperature ranging from 0^0C to 100^0C, various time of dipping viz., 30 seconds, 45 seconds, 60 seconds, 75 seconds, 90 seconds and 120 seconds. The final samples were dried in open air at room temperature for 5 hours and then packed for characterization with mentioning synthesis conditions.

The above process was optimized with 11 Physical parameters viz., 1. Nature (pH) of the dipping solutions (acidic, basic and neutral), 2. Volume of Dipping Solution ranges from 0.5 ml, 1.0 ml, 1.5 ml, 2.0 ml and 2.5 ml, 3. Various types of materials: conventional: varities of plants, trees: Margosa tree , 4. Various Parts of the materials: leaves , 5. Various Dipping solutions: Sodium hydroxide (NaOH), Hydrogen peroxide (H_2O_2), Nitric Acid (HNO_3), Sulphuric Acid (H_2SO_4), Hydrochloric Acid (Hcl), Mineral Water (MW), Salt Water (SW), Double Distilled Water (D2W), Ice Water (Ice W), Hot Water (HW), Pure Water (PW), Ice water mixed double distilled water (IceW+D2W) and Hot water mixed double distilled water (HW+D2W), 6. Various Dipping Timings: 30 seconds, 45 seconds, 60 seconds, 75 seconds, 90 seconds and 120 seconds , 7. Number of Dippings: 1, 8. Various Annealing Temperatures: 600^0C and 800^0C , 9. Various Time of Annealing: 1 minute to 5 minutes, 10. Various Dipping Solution temperatures: 0^0C to 100^0C., 11. Number of Annealing: 1

3. RESULTS AND DISCUSSION

Characterization of CNTs

3.1 Structural Characterization

Margosa tree leaf

Fig. [3.1.1] expresses X-ray diffract gram (XRD) of Margosa leaf annealed at 600^oC (4 minutes) dipped in Hot water mixed with DD water @ 45^oC (volume: 1.5 ml) for 60 seconds shows amorphous (non-crystalline) nature. Open air dry of fresh leaf after taken out from field and annealing on Margosa tree leaf at 600^0C (4 min) leads to complete dryness of leaf. Due to that dehydration (removal of H_2O molecules from C-H-O matrix) takes place in the leaf. Finally it allows carbon atoms only present in the leaf. After annealing, we have to immediately undergone that leaf by dipping into 100^0 C Hot water mixed with DD water (60 Seconds). Due to sudden cooling, the carbon lattices present in the leaf were cracked, split up into individual atoms. Finally that lattice formation is disappeared, leads to amorphous (non-crystalline) nature

3.2 Compositional Characterization

Margosa tree leaf

Fig.[3.2.1] indicates EDAX Spectrum of a 800^0C (5 minutes) heated Neem (Margosa) tree leaf dipped in 100^0 C Hot Water with Double distilled water (volume: 0.5 ml) (2 minutes). EDAX studies on Margosa tree leaf annealed at 800^0C (5 min) dipped in 100^0 C Hot water mixed with DD water (2 minutes) shows compositional elements present in the Margosa tree leaf. Presence of rich Carbon atoms (1^{st} prominent peak in EDAX spectrum) confirmed that the formed tubes are made up of carbon atoms. Which was authentically shows the formation of CNTs. Due to high temperature annealing, dehydration takes place, H_2 atoms were removed. Which was evidentially shown from the EDAX spectrum (i.e., H_2 not found in the EDAX spectrum). Presence of O_2 atom in the EDAX spectrum evidentially shows that oxygen was injected / feed up during annealing. Presence of (Silicon) Si, (Oxygen) O shows that the leaf was enriched with SiO_2 (Soil) in which the tree was grown. Mg, Al, S, N, K and Ca proved that the roots of tree sucked these essential elements (nutrients) from soil for its growth and stability.

3.3 Surface Morphological Characterization

Margosa tree leaf

SEM studies were carried out with a JEOL JSM-840 operated at 20 KV. SEM Photographs of on Margosa tree leaf annealed at 800^0C (2.5, 3 and 4 minutes) dipped in 100^0 C

Hot water mixed with DD water Solution (30, 45, 60 and 75 seconds) (Fig. 3.3.1 – 3.3.4) shows earliest tubular formation of Self assembled suffled / unordered growth of horizontal SWCNTs (longitudinal view) and middle hallow space (hole) covered with single wall in vertical SWCNTs (Top view). SEM images also revealed the earliest tubular formation of single walled CNTs (horizontal view). The image picture out the tubular formation from earliest cracking of carbon lattices due to sudden cooling, after high temperature annealing on Margosa leaf. The images also depicted the earliest growth of vertical SWCNTs (Top view) and an enlarged view of matured growth of vertical SWCNTs (Top view). The images showed that 100^0 C Hot water mixed with DD water solution acts as a suitable medium (any solution which has pH = 7) for the formation of (suffled) vertical growth of SWCNTs with middle hole surmounted by single wall in Margosa leaf. The images showed tubular structure of matured horizontal SWCNTs as individual fibres exhibit a middle hallow empty space with single wall outer cover in Margosa leaf.

Fig. [3.3.1] emphasized SEM photograph of a 800^oC (4 minutes) heated Neem (Margosa) tree leaf dipped in 100^oC Hot Water mixed Double Distilled Water (HW+D2W) Solution (volume: 2.0 ml) for 45 seconds. It proved pre-final, high grade tubules threading out from carbon core to form horizontal shuffled /unordered Single walled Carbon Nano tubes (SWCNTs). Fig. [3.3.2] explored SEM photograph of a 800^oC (3 minutes) heated Neem (Margosa) tree leaf dipped in 100^oC Hot Water mixed Double Distilled Water (HW+D2W) Solution (volume: 1.5 ml) for 60 seconds. It enumerated the intermediate growth stage of high grade vertical tubules threading out from carbon core with inner hallow space to form vertical SWCNTs (cross sectional view). Fig. [3.3.3] picture out SEM photograph of a 800^oC (3 minutes) heated Neem (Margosa) tree leaf dipped in 100^oC Hot Water mixed Double Distilled Water (HW+D2W) Solution (volume: 1.5 ml) for 75 seconds. It picture out the intermediate growth stage of high grade vertical tubules threading out from carbon core with inner hallow space to form vertical SWCNTs (an eagle view of vertical cross section). Fig. [3.3.4] explained SEM photograph of a 800^oC (2.5 minutes) heated Neem (Margosa) tree leaf dipped in 100^oC Hot Water mixed Double Distilled Water (HW+D2W) Solution (volume: 2.5 ml) for 30 seconds. It focused the super final, high grade horizontal tubules maturely grown out from carbon core with inner hallow space to form horizontal SWCNTs (longitudinal view).

3.4 Nanostructural Characterization

Margosa tree leaves

HRTEM studies were carried out with a JEOL JSM-840 operated at 20 KV. In Fig. [3.4.1], HRTEM image emphasized intermediate stage splitting and in-homogeneous scattering of high grade pentagonal, hexagonal carbon matrices/networks during cooling process from

800^0C (3 minutes) red-hot Margosa tree leaf when dipping in NaOH solution(volume: 1.5 ml) for 60 Seconds based on single time annealing and dipping process. In Fig. [3.4.2], HRTEM image revealed intermediate stage splitting and in-homogeneous scattering of moderate grade pentagonal, hexagonal carbon matrices/networks during cooling process from 800^0C (3 minutes) red-hot Margosa tree leaf when dipping in Ice water mixed double distilled water (IceW+D2W) (volume: 1.5 ml) for 75 Seconds based on single time annealing and dipping process.

In Fig. [3.4.3], HRTEM image revealed intermediate stage coagulation, splitting and in-homogeneous scattering of low grade pentagonal, hexagonal carbon matrices/networks during cooling process from 800^0C (3 minutes) red-hot Margosa tree leaf when dipping in double distilled water (D2W) (volume: 2.0 ml) for 75 seconds based on single time annealing and dipping process. In Fig. [3.4.4], HRTEM image flashes out final stage coagulation, splitting and in-homogeneous scattering of low grade pentagonal, hexagonal carbon matrices/networks during cooling process from 800^0C (1 minute) red-hot Margosa tree leaf when dipping in mineral water (MW) (volume: 0.5ml) for 120 Seconds based on single time annealing and dipping process.

In Fig. [3.4.5], HRTEM image illustrated intermediate growth stage very poor coagulation of low grade pentagonal, hexagonal carbon matrices/networks during cooling process from 800^0C (3 minutes) red-hot Margosa tree leaf when dipping in mineral water (MW) (volume: 1.5 ml) for 75 Seconds based on single time annealing and dipping process. In Fig. [3.4.6], HRTEM image revealed intermediate stage poor coagulation of moderate grade pentagonal, hexagonal carbon matrices/networks during cooling process from 800^0C (2 minutes) red-hot Margosa tree leaf when dipping in Ice water mixed double distilled water (IceW+D2W) (volume: 1.0 ml) for 75 Seconds based on single time annealing and dipping process. The effects of optimizations parameters like pH of the various dipping solutions (acidic, basic and neutral), volume of dipping solutions, various types and parts of the materials, various dipping timings, number of annealing and dipping, various annealing temperature, various time of annealing and various dipping solution temperatures on structural, compositional, surface morphological, nano-structural characterizations of materials and on high grade SWCNTs growth with high yield were studied intensively. Parameters, as inferences from above characterizations were calculated, tabulated and graphically emphasized. Correlation studies between these characterization inferences (such as grain size, purity) and optimization parameters were carried out with a high light on yield of high grade SWCNTs. Beyond all of these, we have carried out a novel feasibility study at first time, which comprises the possible usage of precursor organic carbon sources i.e., margosa (neem tree) leaves for high quality SWCNTs with high yield via a low cost technique and methodology as value in commercial efforts.

4. CONCLUSION

In this over all study, a feasibility study was carried out on margosa leaves under conventional materials category towards possible synthesis of high quality SWCNTs. Also any Dipping Solution (of Volume: 2.5 ml) having pH=7, i.e., neutral solution (normally maintained at room.temp.,RT) (with 30 Seconds as optimum time of dipping) act as an optimum medium which provides suitable environment for high quality, large quantity SWCNTs growth. Similarly 800^0C (having annealing time: 3 minutes) provides suitable background thermal history for high quality, large quantity SWCNTs growth based on single time annealing and dipping process. High purity precursor material yield high grade SWCNTs.

Margosa leaves.

Fig. [4.1] defined the Relation between Quantity of SWCNTs formation (%) and Dipping solution with an over view on Quality grades of SWCNTs grown from Margosa leaves. The graph showed that 100^0 C Hot water mixed with DD water solution acts as a suitable medium (any neutral dipping solution which has pH = 7) for the formation of high quality SWCNTs in Margosa leaf than basic solution which has pH > 7. All of these works have value in nanotechnology, nano-materials processing and device fabrication efforts either as a technical or scientific basis, also as a contribution to the present day state of the art of nano paper battery like energy storage system.

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